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Availability, Uptake and Translocation of Plutonium within Biological Systems: A Review of the Significant Literature

Environmental Monitoring and Support Lab.

April 1976

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AVAILABILITY, UPTAKE AND TRANSLOCATION OF PLUTONIUM WITHIN BIOLOGICAL SYSTEMS:

A Review of the Significant Literature

Environmental Monitoring and Support Laboratory
Office of Research and Development
U.S. Environmental Protection Agency
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This report is a selective review of the literature on the availability of plutonium in the environment and its cycling throughout representative biological systems ranging from large biomes covering hundreds of miles to the molecular transformations within individual cells. No attempt was made to develop a comprehensive bibliography. Rather, references were selected for inclusion as representative documentation for the vast spectrum of material that is available on the subject.

Important general references are listed separately. Thereafter the literature is described in essay form on a subject basis. References cited by number in the text are listed in complete bibliographic form at the end of the report together with an author index. The majority of the material reviewed is limited to relatively recent publications.

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AVAILABILITY, UPTAKE AND TRANSLOCATION OF PLUTONIUM WITHIN BIOLOGICAL SYSTEMS: A Review of the Significant Literature

by
Anita A. Mullen and Robert E. Mosley
Monitoring Systems Research and Development Division
Environmental Monitoring and Support Laboratory
Las Vegas, Nevada 89114

U.S. ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF RESEARCH AND DEVELOPMENT
ENVIRONMENTAL MONITORING AND SUPPORT LABORATORY
LAS VEGAS, NEVADA 89114

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ABSTRACT

This report is a selective review of the literature on the availability of plutonium in the environment and its cycling throughout representative biological systems ranging from large biomes covering hundreds of miles to the molecular transformations within individual cells. No attempt was made to develop a comprehensive bibliography. Rather, references were selected for inclusion as representative documentation for the vast spectrum of material that is available on the subject.

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CONCLUSIONS

This review has briefly covered many topics but has addressed only a small portion of the articles published over the past 25 years.

As additional energy requirements increase the number of nuclear power generators, processing and reprocessing plants throughout the world, the potential increases for the distribution of plutonium within a variety of biomes previously only touched by worldwide fallout. At this time it appears that more is known of the interactions of plutonium within a desert ecosystem than in any other system. The largest single group studying plutonium in the environment is the Nevada Applied Ecology Group (NAEG) which is supported by the Energy Research and Development Administration. Its main concern has been the area of the Nevada Test Site and its immediate environs.

Additional studies are needed to determine the physicochemical interactions of plutonium in areas receiving moderate to heavy rainfall, which results in lush vegetation with its attendant litter and greater quantity of decomposition products affecting soil chemistry and fertility. Temperature extremes causing seasonal variations in plant constituents, endemic and transient wildlife populations, invertebrate community size and average age of individuals within these ecosystems all must be taken into consideration.

Differing soil types are known to affect the translocation and availability of plutonium. What is not clearly defined is the effect of adding amendments to the soil in order to increase fertility. Nitrogen and trace minerals may be added to soil in many chemical forms, each of which may change the availability of plutonium for absorption by flora or fauna.

The selectivity of organisms for iron and plutonium present in the same biomass is not thoroughly understood. This may be affected by seasons, nutritional needs, altitude and age of the receptor.

Investigations underway by NAEG indicate that the availability of plutonium may be increased each time it is ingested by an animal, passes through the digestive processes and is excreted. This may concentrate plutonium in vegetation growing on plots of ground previously fertilized by the application of contaminated manure. More work must be done before these complex interactions can be understood.

Some work has been completed on the behavior of plutonium in aquatic systems. The reactions of plutonium particulates in fresh and saline water are just beginning to be understood. Incorporation of particulates within bottom sediments may isolate plutonium and prevent dissolution for long periods of time but violent agitation, e.g., storm wave action, mechanical mixing or erosion and flooding of large land areas and deposition of the sediments on farm lands may cause a release of this stored plutonium and increased uptake by biological systems. This mechanism should be better understood since most nuclear power plants are located near large sources of water increasing the potential for low level chronic release in effluent streams.

The role microorganisms play in affecting availability of plutonium in soil-plant relationships is being studied on a limited basis. Little is known of plutonium interaction with microorganisms present in the gastrointestinal tract. Work has started on the absorption of plutonium by algae. Ingestion of this algae by aquatic organisms may lead to further concentration and transfer up the food chain.

There are many studies underway by the NAEG as summarized by Dunaway and White[24] in 1974. Many of these studies seek to further information on the behavior of plutonium in fauna found within a desert environment. These studies encompass tissue concentration of plutonium in cattle grazing in contaminated areas, rodents and lizards inhabiting the same area, transfer of various forms of plutonium to milk by dairy cows, and concentration of plutonium in tissues and eggs of chickens ingesting differing forms of plutonium.

Many authors have devoted much time to extrapolating the results obtained with laboratory animals to man. This comparison though perhaps valid in many instances, may prove to be of limited use when the complicating factors and discriminating mechanisms inherent in man's physiology are taken under consideration. The effects of chronic plutonium contamination in humans necessary for a true evaluation of its hazards will continue to be a source of conjecture for many years in the future. Man can only hope to limit the concentration of this potentially hazardous radionuclide in the environment until additional knowledge is forthcoming.

INTRODUCTION

OBJECTIVE

The objective of this review is not to develop a comprehensive bibliography on plutonium, but rather to identify and comment upon the significant and relevant literature representing the many detailed studies of the interaction of plutonium in biological systems. Material included in this review has been selected to provide an overview of the subject and to indicate where deficiencies may exist in biological data.

Plutonium, produced for the first time in 1941, has been released and distributed throughout the biosphere during the last 25 years. The physicochemical changes induced by natural processes may enhance the biological availability and increase the concentration of plutonium in specific biomes thereby causing greater biological effects and risk to mankind.

The concern then is to limit the production and control the use of plutonium until such time as the very complex interactions with biological systems have been investigated and a better understanding of potential hazards has been achieved. The need for energy makes it imperative that mankind make most efficient use of available sources and channel the potential of plutonium-derived energy to the benefit of all, while minimizing the deleterious effects on man and his environment. Only by an overall understanding of what has been found in the past can guidelines be set for the future.

APPROACH

Literature included in this report was selected on the basis of its significance and relevance from a variety of bibliographies, general references and abstracts. Where the original reference was not available to judge the value of the reference, the abstract was consulted to determine its applicability.

ORGANIZATION

This report lists annotated bibliographies, important general references and abstracts. Thereafter, the literature is reviewed in essay form on a subject basis. The subject headings are covered in individual sections and under each of these, several subsections review literature on a given topic. The 419 references reviewed are cited in the appropriate subject divisions of this report and in complete bibliographic form at the end of the report in the order mentioned in the text. Finally, an author index of the references cited is included to facilitate the location of particular material.

BIBLIOGRAPHIES

The following bibliographies are closely related to the subject.

(1) Environmental Aspects of Plutonium - A Selected Annotated Bibliography, September, 1972, Environmental Plutonium Data Base Group, Oak Ridge National Laboratory, Oak Ridge, TN, ORNL-EIS-72-21, 387 p.

(2) Ibid (Suppl. 1) August, 1973, ORNL-EIS-73-21, 479 p.

(3) Ibid (Suppl. 2) February, 1974, ORNL-EIS-74-21, 272 p.

These three volumes contain over 2,000 references on the environmental aspects of plutonium and uranium at the Nevada Test Site (NTS). The literature was selected to meet the needs of the Nevada Applied Ecology Group (NAEG) of the Nevada Operations Office, Las Vegas, Nevada.

(4) Environmental Aspects of the Transuranics - Compiled and edited by Martin, F. M., Sanders, C. T., and Talmage, S. S., Oak Ridge National Laboratory, TN, ORNL-EIS-74-2 (Suppl. 3) December 1974, 226 p.

This bibliography contains 528 references on the environmental aspects of uranium and the transuranic elements, with a preponderance of material about plutonium. Indexes are given for authors, subjects, categories, keywords, geographic locations, permuted titles, taxons, and publication descriptions.

(5) Biological Effects of Plutonium. A Bibliography - Compiled by Suratno, P., CE-BNL-Bib-6, 60 p.

This bibliography was compiled mainly from Nuclear Sciences Abstracts for the period 1969-1973. It was produced in support of the continuing work at Berkeley Nuclear Laboratories on the health physics problems associated with the transuranic elements. Its arrangement is in three sections: A. Biological Effects (47 refs); B. Physiological Aspects (110 refs); and C. Techniques of Measurement (60 refs).

General Reference Reviews and Summaries

The following reports or references give reviews and summaries of available information related to the subject.

(1) Uranium-Plutonium-Transplutonic Elements, Hodge, H. C., Stannard, J. N., and Hursh, J. B., Eds., Springer-Verlag, New York, 995 p., 1973.

This book is separated into three broad categories and 21 chapters which deal separately with uranium, plutonium, and the transplutonic elements. In general the sections discuss history; chemical and physical properties; distribution, excretion, and effects; body burdens; bioassay; health physics aspects; and environmental considerations.

(2) Radiobiology of Plutonium, Stover, B. J. and Jee, W. S. S., Eds. The J. W. Press, University of Utah, Salt Lake City, Utah, 552 p., 1972.

This book is separated into 25 chapters dealing with general topics of historical background, metabolism and effects of plutonium as pertains to beagle studies, metabolism of ^{239}Pu in rodents, use of chelating agents, and plutonium in man.

(3) The Metabolism of Compounds of Plutonium and Other Actinides, ICRP Pub. 19, Pergamon Press, New York, 59 p., May, 1972.

This report reviews literature up to 1972 relating to the chemistry of plutonium as related to biological behavior, e.g., entry of plutonium by inhalation, absorption and retention of deposited actinides. Three tables are given which summarize principal inhalation studies with plutonium compounds in experimental animals, absorption of actinides from the gastrointestinal tract and distribution of actinide compounds in experimental animals and man.

(4) "A Review of Transuranic Elements in Soil, Plants, and Animals," J. Environ. Quality 2:1, pp. 62-65, 1973.

Published information concerning the distribution and fate of neptunium, plutonium, americium and curium in terrestrial ecosystems is reviewed in this article.

(5) "Physiopathology of Plutonium Contamination: Fundamental Concepts," Saenz, M. D. L. and Ramos, E., translated by Ralph McElroy Co. for Los Alamos Scientific Laboratory, Los Alamos, LA-TR-74-16, 40 p., 1973, translated 1974.

Published information concerning the physiopathology of plutonium was reviewed including internal contamination, effects, dose and therapy.

(6) "Consideration of Reactor Accident Exposure Guides for Plutonium," Bair, W. J., BNWL-SA-4968, 23 p., 1974.

This report reviews some of the biological aspects needed to be considered in establishing an emergency reference dose for plutonium. The discussion is limited to plutonium entering the body through the respiratory tract.

(7) "Evaluation of Plutonium-Contamination in Radioactive Waste Disposal Areas with Respect to Their Potential Hazard and Eventual Disposition," McCurdy, D. E., M. L. Wheeler, M. D. McKay, and R. K. Lohrding, Quarterly Report Transuranic Solid Waste Management Research Programs October-December 1973, Los Alamos Scientific Laboratory, p. 17-55.

This report evaluates the plutonium contaminated waste disposal areas of the Los Alamos Scientific Laboratory (LASL) and analyzes the risk of natural and man-made disasters that could be causal events for releasing stored waste to the environment, the resulting abiotic transfer processes for movement of the released plutonium-contaminated waste, biological transport mechanisms, radiological assessment of the intake of transuranic elements and the radionuclide inventory of waste disposal areas at LASL are also discussed.

(8) The Dynamics of Plutonium in Desert Environments, Dunaway, P. B. and M. G. White, Eds., NVO-142, 369 p., July, 1974.

This is a progress report of projects of the Nevada Applied Ecology Group, U.S. ERDA, Nevada Operations Office and contains summaries of work presently underway as well as preliminary reports of completed projects. The several topics covered are soils, statistics, vegetation, animals, resuspension, distribution, and inventory modeling and information as applies to plutonium on the Nevada Test Site and surrounding environs.

POTENTIAL SOURCES OF PLUTONIUM

GENERAL

Plutonium is a radioactive element of increasing importance as the need for energy sources becomes more acute. It is one of the most toxic radionuclides because of its biological behavior as well as its physical characteristics.

As summarized by Saenz and Ramos[1] in 1973, the importance of plutonium has been apparent since it was produced for the first time by Seaborg and others as reported by Weast[2] in 1966. Although the existence of plutonium in nature has been proven, its natural quantities are so low (Levine and Seaborg[3] in 1951 and Hoffman[4] et al. in 1971) that the risk comes from its artificial production.

Of the radioactive plutonium isotopes, ^{239}Pu is the one most used in the nuclear industry where it is produced by irradiation of ^{238}U in nuclear reactors. Its high specific activity and long half-life determine its high radiological toxicity (Saenz and Ramos[1]).

Increasing use of plutonium in nuclear fuel, industrial, biomedical and space applications, as well as weapons development, increases the possibility of accidental releases contaminating the biosphere.

Since 1952, weapons tests and nuclear accidents in space have released plutonium into the biosphere with resultant fallout over the entire earth (according to the estimates of Cherdyntsev[5,6] et al. in 1968 and 1970) approximating a concentration of ^{239}Pu of 10^{-12}% when mixed in the first few centimeters of soil. The values of liberated plutonium given by the Scientific Committee of the United Nations and reported by Norwood[7] in 1971, assumes an activity of 0.5 mCi of ^{239}Pu, equal to eight tons.

Global air activity values for ^{239}Pu were shown to be 10^{-15} Ci/m^3 of ^{239}Pu in 1963 as reported by DeBortoli[8] et al. in 1966 and 10^{-17} to 10^{-16} Ci/m^3 in the USA in 1965 as calculated by Drobinski[9] et al. in 1966. These values appear to coincide in order of magnitude with later maximums arrived at by Volchok and Kleinman[10] in 1971.

Although the risk from weapons testing would appear to be low, the increasing use of plutonium in industry, medicine and aeronautics has caused Seaborg[11] in 1970 to forecast that these applications will use 60 to 80 tons of ^{239}Pu in the next few decades and up to 6 tons of ^{238}Pu by the end of the century.

Environmental contamination as the result of accidents was reviewed by Eisenbud[12] in 1973. The events discussed include: fallout from the thermonuclear detonation of March 1, 1954; the accident to the Windscale Reactor number one of October 1957; the Oak Ridge plutonium release of November 1959; the Army Stationary Low-Power Reactor (SL-1); the Houston incident of March 1957; the Fermi fuel meltdown; the abortive reentry of the SNAP 9-A; and the plutonium fire at Rocky Flats.

WEAPONS DEVELOPMENT

The first formal environmental contamination experiments with plutonium were carried out under the auspices of the U.S. Atomic Energy Commission (AEC) at the Nevada Test Site. In 1956 a test (Project '56) was carried out to study the behavior of released plutonium in air, soil, and the desert environment. No biological work was included in this experiment. As reported by Stannard[13] in 1973, "Operation Plumbbob" conducted in 1957, a full scale biomedical program was undertaken by a group from the University of Rochester (Wilson[14] et al.) along with more elaborate studies of aerosols, cloud physics, decontamination and area monitoring studies by a group from Sandia Corp. (Cowan[15]) and the United States Air Force. The levels of ground contamination were on the order of 2.6, 40 and 560 micrograms per square meter of desert soil.

"Operation Roller Coaster" was a joint United States-United Kingdom venture and was carried out in a remote area of north central Nevada. There were four test firings and the emphasis was placed on inhalation of plutonium from cloud passage according to Wilson and Terry[16,17] in 1965 and 1968.

An environmental inventory of the Los Alamos area was reported for the period July 1, 1972 to March 31, 1973, by Hakonson[18] et al. in 1973. Included in this survey was the Trinity area, scene of the world's first nuclear detonation (July 1945). A similar study of the 2,200 hectares surrounding Los Alamos was reported by Johnson[19] in 1972. These environmental assessments included careful search of the administrative records to determine the extent to which the land might have been used or involved in the Laboratory's activities. Extensive measurements of the radiation levels in the field, and radiochemical analysis of numerous soil and vegetation samples were completed. Results showed that all measured values were comparable to reported worldwide levels, and no radiation or radioactive contamination observations were encountered that are of radiological health or environmental concern. In a similar report by Hakonson and Johnson[20] in 1973, a comparison was made between the data collected 20 years earlier and that collected in 1972. The major change observed was an increased migration of plutonium downward through the soil profile. Concentrations of plutonium in vegetation and rodents were too low to make valid comparisons.

The comprehensive review prepared by Eisenbud[21] in 1973 of fallout distributed from nuclear explosion includes the amounts and distribution of radioactive debris produced in weapons tests; methods of estimating how radioactive debris is partitioned among the three components of fallout (the fallout in the immediate vicinity of the explosion, the debris injected into the troposphere, and the debris injected into the stratosphere); and the behavior of plutonium released in weapons tests.

In the quarterly reports of the New York Operations Office Health and Safety Laboratory, Hardy[22,23] tabulates the data from the fallout monitoring program. The plutonium content of atmospheric samples obtained through Project Air Stream during 1970 and 1971 and the plutonium content of stratospheric samples obtained through the High-Altitude Balloon Sampling Program during 1969 and 1970 are included in the report for March-June 1971. The June-September 1972 report shows plutonium deposition at worldwide land sites and those collected at Atlantic Ocean weather stations.

Environmental surveys of the main areas of plutonium deposition at the Nevada Test Site are included in the Nevada Applied Ecology Group Progress Report edited by Dunaway and White[24] in 1974.

WEAPONS IN TRANSPORT

American bombers carrying nuclear weapons have crashed in Spain in 1966 and Greenland in 1968. Problems connected with the detection and decontamination of these areas were discussed by Hvinden[25] in 1973. The area contaminated in Spain was cleared sufficiently after 2 or 3 months to allow agricultural use. In Greenland, measurements of fish, birds, foxes and seals showed no contamination above allowable limits.

In a follow-up study, Hanson[26] in 1972 was able to detect small amounts of ^{239}Pu and ^{240}Pu in lichens collected near the Greenland site during July-August of 1968. This activity was associated with particles estimated to be 0.5 to 1.0 micrometers in diameter containing slightly greater than background levels of plutonium radioactivity.

FUEL PROCESSING AND REPROCESSING

Increasing utilization of nuclear fuels will result in wide-scale plutonium recovery processing, reconstitution of fuel, transportation and extensive handling of this material. According to Paxton[27] in 1972, there have been only six supercritical accidents spread over 20 years of processing fissile material. None of these was associated with mechanical processing, storage or transportation. All occurred in recovery plants and all occurred with aqueous solutions; four involved highly enriched uranium, and two involved plutonium. One of those involving plutonium occurred at the Los Alamos Scientific Laboratory - December 30, 1958, and the other at the Recuplex Plant at Hanford, Washington, April 7, 1962. The results of these 6 accidents have been 2 deaths, 19 significant overexposures to radiation, no equipment damage, and negligible loss of fissile materials. In no case was there any danger to the general public.

Many safeguards are built into facilities used for processing of nuclear fuels so that confinement of radionuclide release to the environment is kept to a minimum. It is a well recognized problem that plants handling long-lived radionuclides may release extremely low levels of radionuclides daily through filters or other pathways. Although these releases may not be generally measured in terms of air concentration, after many years of operation small quantities of the material can be detected outside of the facility. This has happened at Rocky Flats and other areas according to Biles[28] et al. in 1971.

A study was made of the possibility of hazardous environmental plutonium releases caused by uncontrolled oxidation of plutonium within the enclosure at Rocky Flats (Hunt[29]). The report reviews the existent observed data on restricted plutonium release and then constructs a release model relating the free-release source strength (i.e., the source strength outside the enclosure) and the parameters describing the release. Based on both the observation data and the model calculations, the conclusion follows that restricted release is unlikely to lead to dangerous free-releases of a plutonium aerosol. The model calculations specifically show that the maximum credible accident assumed, would be safe by about 2 to 5 orders of magnitude. However, the calculations indicate that maximizing either the plutonium release rate or the asymptotic temperature reached in an enclosure can cause a free-release exceeding the safe rate.

FIRES

The pyrophoric nature of plutonium metal has led to several plutonium fires. The major fires occurred at Rocky Flats in 1957 and 1969 and resulted in airborne plutonium being distributed over a wide area surrounding the facility and resulted in contamination of the soil as reported by Hardy and Krey[30] in 1970.

An analysis of 46 incidents involving plutonium metal or compound under thermal stress has been reported by Mishima and Schwendiman[31] in 1971. These incidents, occurring in the nuclear industry during the period from 1952 to 1967, are cataloged as to the nature of the material involved, the initiating event and the consequences.

WASTE DISPOSAL FROM PLANTS

For several decades, transuranic-containing solid waste has been placed in shallow burial grounds at several of the AEC contractor facilities. The hazards of the release of this radioactivity from the waste burial environment at the Los Alamos facility was discussed by McCurdy[32] et al. in 1974. The possible catastrophic events leading to the release of these materials are addressed and a model is proposed for the movement of these radioactive materials throughout the environment.

A review of processing of plutonium-contaminated liquid wastes at Los Alamos was reported by Emelity and Christenson[33] in 1971. This report lists the quantities of liquid wastes processed during the years 1966 through 1970.

Over the past 20 years, the National Reactor Testing Station (NRTS) in Idaho has accepted radioactive waste for burial. Wastes received included materials contaminated with transuranic elements, primarily from AEC's Rocky Flats operation, as well as items contaminated with fission products and neutron activated materials from various onsite operations. Early disposal methods varied from stacking of drummed and boxed waste into pits to random dumping of various categories of waste into common trenches or pits. Reassessment of past disposal practices and concern for long-term isolation of transuranic wastes led to the establishment of above-ground retrievable storage in 1970. However, about 54,000 cubic meters of alpha waste remains buried at NRTS. The plan for retrieval of this waste for treatment and storage in a Federal depository was reported by Hickman[34] in 1974.

Liquid wastes from the plutonium finishing plant at Richland, Washington, have been discharged to subsurface disposal facilities (enclosed trenches) since the startup of the facility approximately 22 years ago. The removal and disposal of this contaminated soil is discussed in AEC Environmental Statements[35,36] of January and April of 1972.

New guidelines for the interim storage of solid transuranic wastes were proposed by the H-Division staff of Los Alamos[37] in 1974. A summary of current practices in solid waste disposal is included.

A review of radioactive waste disposal and its eventual pollution of groundwater from this waste was completed in 1974 by Todd and McNulty[38].

SHIPMENT

It has been estimated by the U.S. Atomic Energy Commission[39] in 1972 that by the year 2000 the total nuclear power generating capacity in the United States will be approximately 1200 gigawatts. Over the next 25 years, the expansion will increase the need for shipping ^{239}Pu (produced in light water reactors and liquid metal fast-breeder reactors) from fuel reprocessing plants to fuel fabrication plants where these fissile isotopes can be reconstituted into reactor fuel elements.

If aqueous solvent extraction processes are employed for separating plutonium in the spent fuel, the plutonium will emerge in the reprocessing plant product streams as nitrates in aqueous solutions. The problems and hazards inherent in shipping these solutions were discussed by Ulrich[40] in 1974. In a conference on transportation of nuclear material, Wischow[41] in 1970 reported case histories of misrouted and misplaced shipments. Corrective action and regulations pertaining to physical prohibition of fissile materials were briefly discussed. Hazards associated with transportation accidents in which plutonium was subjected to gasoline fires were reported by Mishima and Schwendiman[42] in 1973.

Becker[43] in 1971 reported on steps that LRL was taking to improve controls over packing of plutonium shipments. Two case histories were cited. The importance of quality control in packaging $Pu(NO_3)_4$ solution and PuO_2 powder was discussed in an article by Brown and Heaberlin[44] in 1974. An industry-wide survey was conducted of 775 shipments involving 6,200 packages over a period of 3 to 4 years. Package closure error could result in breaking of containment during shipment or a series of errors might release the contents during normal shipment. A listing of closure faults is provided with the number of times the faults have been observed.

This is not a complete record of these faults as records on errors were not kept consistently as required to be kept. Although increased quality control procedures implemented since 1972-1973 have decreased the frequency of these errors, they have not been entirely eliminated.

The problems of calculating risk of criticality in transporting fissile materials were addressed by Thomas[45] in 1971. Proper packaging and container sizes are discussed.

NUCLEAR REACTORS AND RADIOISOTOPIC GENERATORS

Plutonium is produced by irradiating uranium with neutrons in a nuclear reactor. Its isotopic composition depends on the time in the reactor and the type of reactor. This plutonium may be recycled in light-water reactors. Utilization in this way will require reprocessing and fuel refabrication.

The introduction of commercial fast-breeder reactors is not expected to occur until the mid-1980's according to Pigford and Mann[46] in 1973. The fast-breeder reactor has excellent fuel economy. It is fueled with plutonium and "breeds" plutonium from ^{238}U, creating a surplus for the first fuel charge of other fast-breeder reactors. The danger from this type of reactor, according to Hamilton[47] in 1971 is that the chain reaction can easily run away, creating a supercritical mass which would eventually be blown apart. The total radioactivity in the yearly amount of fuel discharge, at the time of discharge, is greater for the fast-breeder reactor than for the light-water plant because of the need for more frequent replacement of core fuel in the breeder. The yearly amount of plutonium in the fast-breeder fuel cycle is about eight-fold greater than in the light-water nuclear plant. An experimental 1000 Mw(e) fast-breeder reactor will contain about 1.8 to 2.8 tons of plutonium corresponding to 2.2 to 3.5 million curies of plutonium according to the U.S. Atomic Energy Commission[48] report of 1972. A flow sheet listing the material and environmental release for a breeder reactor nuclear power plant was presented by Pigford and Mann[46] in 1973.

A historical résumé of the breeder reactor program and its impact on the environment was given in a report by Daub[49] in 1972. A program plan listing fuel materials and accident conditions for a liquid metal fast-breeder reactor (LMFBR) was presented by the Argonne National Laboratory[50] in 1972.

Safety aspects of nuclear fuel cycles were reviewed by Wymer[51] in 1971. Future developments and requirements for the nuclear fuel cycle are also discussed. Plutonium utilization in boiling water reactors is summarized in a report by General Electric Company,[52] released in 1971. Activities associated with the AEC-sponsored Integrated Safeguards Experiment and Plant Instrumentation Program are reported. A review of plutonium utilization in thermal reactors was presented by Schmid[53] in 1973. Recycling, fueling, decontamination and safety are discussed. The drawbacks of nuclear power were discussed by Engstroem[54] in 1972. Siting of power plants, radiation hazards and environmental impact are included. The dangers associated with nuclear power reactors were described by Lapp[55] in 1975. The possibilities of accidents occurring are presented in an analytical diagram called an "accident tree." Of all the possibilities, the probability of a few hundred of the most important was determined and of these 15 were judged to be the major sources of risk. An assessment of accident risks in U.S. commercial nuclear power plants was reported by Rasmussen[56] in 1974.

The health physics aspects of fast-breeder reactors were discussed by Heine and Hart[57] in 1971. The specific aspects are grouped according to segments of reactor operations. Under certain hypothetical accident conditions where fuel vaporization could occur, plutonium could be the dominant hazard.

The results of a radiological environmental survey in the vicinity of EBR-II were given by Oltman[58] et al. in 1973. Measurements were made at radial distances up to 5 miles from the effluent stack. Soil samples were obtained and analyzed for plutonium content. No plutonium deposition pattern could be constructed.

A series of studies to evaluate the fractional airborne release of plutonium under various postulated reactor accident conditions was completed by Mishima and Schwendiman[59] in 1972. Data generated in earlier laboratory scale studies were also reviewed. Topics dealing with the acceptability of nuclear fission for energy sources were discussed by Gofman[60] in 1972. The biological problems associated with nuclear fission, the fission product problem, the plutonium problem and the major accident problems are included. The University of Washington nuclear reactor plutonium contamination incident of June 13, 1972, was discussed by Hilborn[61] et al. in 1972.

Both major and minor accidents involving the release of aerosols of sodium are possible during the generation and maintenance of a fast reactor. These aerosols could be radioactive, involving not only the radioactive sodium, but also fission products, uranium and plutonium. This problem was reviewed by Kotrappa[62] in 1973. The use of nuclear power generators, batteries and heat sources for use on land, in space vehicles, ocean diving equipment and for powering artificial pacemakers, organs, and other medicinal uses increase the possibility of environmental contamination from these sources. A detailed nuclear safety evaluation of the SNAP-19 C generator was conducted by Conway and Dobry[63] in 1973. In 1972 Weiner[64] reported on the safety testing of a ^{238}Pu fueled Radioisotope Thermoelectric Generator (RTG) for the Navy Transit Satellite. The containment of the ^{238}Pu fuel was checked for all normal reentry environments and for all accident environments during prelaunch, launch pad, early launch abort and pre-orbital insertion abort.

The operating experience and advances in the safety of space nuclear power systems fueled with ^{238}Pu were reviewed by Dix[65] in 1972. The operational safety experience with eight NASA launches of spacecraft powered with nuclear energy since 1968 including two Nimbus weather satellites, the Apollo 12, 13, 14, 15 and 16 lunar landing missions, and the Pioneer 10 Jupiter mission was discussed. The concept of isotopic power generators is explained in a 1971 article by Penn[66]. The heart and pacemaker batteries and the UKAEA RIPPLE (radioisotope-powered prolonged life equipment) generator is described. In 1971 Williams[67] developed models for estimating the extent to which radioisotopic fuel forms will vaporize if released to the fires resulting from the catastrophic aborts of rocket booster vehicles. Pad aborts of both liquid- and solid-propellant systems are considered and aborts after lift-off are also treated. Estimates are given for the size and activity spectra of the radioactive particles produced when the vapor recondenses. Numerical results are specifically calculated for certain $^{238}PuO_2$ fuel forms (microspheres, solid-solution cermet, and plutonia-molybdenum cermet). Tables for the ingrowth of daughter products from the decay of plutonium isotopes used in heat sources are given in an article by Haas and Campbell[68] in 1972. The typical composition of the plutonium metal used in the fabrication of the sources is also listed.

The general considerations of radioisotopic power and its international aspects were discussed by Cellini and Stadie[69] in 1972. The work of the OECD Nuclear Energy Agency since 1967 in studying radioisotope batteries, particularly for cardiac pacemakers is discussed. This work includes heat source development, capsule evaluation, safety assessments of radioisotope heat sources under normal and accident conditions, radiation hazards evaluations, and human implantation of ^{238}Pu fueled pacemakers which began in April 1970.

Safety tests of plutonium fueled devices for medical applications were presented by Warner[70] in 1972. Results demonstrate that plutonium devices are suitable for widespread implantable medical applications. The current status of radioisotope-powered artificial organs was given in 1972 by Toyoshima and Hart[71]. The basic problems of energy sources for implant-type organs and the status of high reliability long-life (10 years) ^{238}Pu power sources were discussed.

The use of radioactive material as a power source implanted in the human body such as ^{238}Pu batteries to drive cardiac pacemakers, becomes more important. In most cases the amount of radioactive material implanted in the body is considerable and the nuclide hazardous (Hunzinger[72]). Bearers of isotopic batteries are not necessarily old people bound to hospital beds. They include all ages and persons in their full professional activity. A bearer of an isotopic battery can hardly be confined to a controlled zone, the establishment of which is a well known principle of control in radiation protection, nor can the person be treated like a consignment of radioactive material and be subject to the well established test procedures for these packages. The safety principles of isotopic sources implanted in a freely moving human are outlined in this paper. The focus is on the exposure of other people to penetrating radiation, on partial loss of the radioactive material, i.e., exposure to contamination and the total loss of control over the source. Safety in industrial fabrication of implantable devices powered by radioisotopes was discussed by Matheson[73] in 1971.

TRANSPORT OF PLUTONIUM FROM SOURCES TO BIOSPHERE

INTRODUCTION

The presence of free plutonium in the atmosphere, ground and water can lead to direct contamination by inhalation or through ecological cycles. Understanding the transport mechanisms for the various physical and chemical forms of plutonium from the contaminating source to the biosphere is complicated by the nature of plutonium chemistry. The fate of the released plutonium is uncertain and complex.

Studies have been conducted by several investigators to determine the problems associated with plutonium cycling in the environment and reported in 1973 by Carfagno and Westendorf[74]. These authors discuss the research being done to determine the mobility of radionuclides in major segments of terrestrial environments. Reciprocal transfers of environmental radioactivity are investigated for three major components--soil, plant, and animal.

The environmental levels of plutonium at U.S. Atomic Energy Commission installations were summarized in a report by the U.S. Environmental Protection Agency[75] in 1973. The sources and activities of plutonium found in environmental samples are given.

The behavior of plutonium in ecosystems and its biological effects on natural fauna and flora as applied to desert ecosystems at the Nevada Test Site were presented by Romney and Davis[76] in a report published in 1971.

The environmental monitoring program for a nuclear fuel reprocessing plant (located in the South Carolina coastal plain), as based on a man-environment ecosystem concept, was described by Platt[77] et al. in 1973. The principal pathways to man--atmospheric, terrestrial, and aquatic--are each subdivided into natural, recreational and domestic components. The use of ecologically defined samples provides rates of movement and bioaccumulation that lead to ecological models that are transferrable to other nuclear industries.

Research to provide data on the mobility and transport of plutonium in environments associated with nuclear power production facilities of the humid eastern United States was outlined by Reichle[78] et al. in 1974.

A model for predicting the redistribution of particulate contaminants from soil surfaces was proposed by Travis[79] in 1975. The usefulness of this predictive tool is demonstrated by calculations involving mixture of particulate $^{238}PuO_2$ in highly erodible soils under dust storm conditions. Time dependent surface concentration and breathing zone exposure isopleths, evolving from a small contaminated area, show the potential hazard from wind eroding toxic materials.

Wind distribution of plutonium released by the fires at Rocky Flats was discussed by Poet and Martell[80] in 1974.

CHEMICAL AND PHYSICAL STATES OF ENVIRONMENTAL PLUTONIUM

Since the chemical and physical states of plutonium determine its transport throughout ecosystems, these states will be discussed separately.

Soil

Soil chemistry mechanisms partially control the biotic availability of plutonium over long time periods. The chemical form of radionuclides in soil in the vicinity of Oak Ridge National Laboratory and the character of secondary reaction products and the availability of nuclides to plants after weathering under natural environments was included in the report by Carfagno and Westendorf[74] in 1973.

Miner[81,82] et al. in 1972 and 1973 give the results of studies on the chemical behavior of plutonium in soil-water environments. Plutonium dioxide and soluble plutonium were shaken with a soil-water mixture and the plutonium in the various phases was measured.

Isotopic ratios and chemical constituents of plutonium in soils and sediments of the Argonne National Laboratory were listed in 1974 by Sedlet[83] et al.

The chemical nature of waste solutions discharged to the soil of the Hanford facility and a study of their present form were reported in 1973 by Swanson[84]. The leach behavior of plutonium in these soils was also investigated.

As stated by Krey[85] in 1974 and Poet and Martell[86] et al., the knowledge of the chemistry of fallout plutonium in soil is not very complete. They tabulate the mobility of plutonium in soil from Rocky Flats. Rhodes[87,88] in two articles published in 1957, pointed out the polymerization known to occur in soil. The slow rate of diffusion and leaching of plutonium by groundwater at the Hanford facility was reported by Hajek[89] in 1966. Self-diffusion of plutonium in soils will only occur due to migration of molecular size particles through soil solutions. He concluded that plutonium could be expected to move about 85 mm in 2.4 X 10^5 years by this process. This was predicted by using soils moistened with 20% water content by volume. This is a high water content when considering desert environments.

The rate of transport of plutonium out of a waste burial area by migrating soil moisture is dependent on many factors. The chemical forms of the isotopes and their respective solubility in water must be determined. The amount of dissolution of plutonium from waste is strongly affected by the chemical form of plutonium present, and by the chemistry of the leaching solution. In general, soluble salts increase the solution rates, as do organic complexing agents [Los Alamos Scientific Laboratory (LASL)[90] in 1974]. In 1974, LASL[90] and Patterson[91] et al. commented that plutonium leaching from soil appears to be more affected by solubility than by exchange reactions.

Plutonium absorption by soils is equally complex, being strongly affected by pH, as well as the concentrations of other metallic ions. Plutonium appears to be present as a polymer in most solutions and is highly sorbed by natural materials. In general, it behaves as a cation at pH values up to about 12, at which point it exhibits strong anionic properties.

Dissolution and absorption of plutonium can be quantitatively described in terms of a distribution coefficient (ratio of material concentration on solid phase to concentration in liquid phase, Kd), as in Christenson[92] et al. in 1958. A table showing solution percentage and distribution coefficients for plutonium in waste and soil was given in the LASL[90] report.

The distribution and characterization of plutonium in soils from the Nevada Test Site were reported by Reichle[78] et al. in 1974. The leachability of plutonium from soils containing different particle sizes was also given.

The physical aspects of plutonium in soils affect its solubility and transport by resuspension. Plutonium resuspension from soil is the subject of reports by Michels[93], Sehmel and Lloyd[94,95]. These reports cover resuspension of plutonium from the vicinity of Rocky Flats and investigate effects of particle size and wind speed.

The evaluation of the resuspendable amounts of plutonium-238 in soil from the vicinity of the Mound Laboratory at Miamisburg, Ohio, was given in summary reports by Carfagno and Westendorf[96,97,98] in 1972 and 1973.

Results of analysis of silt samples from the Great Miami River were also given.

Air

The importance of knowing the chemical and physical forms and the uncertainties involved in estimating inhalation exposures from the ^{239}Pu contamination at Rocky Flats were reviewed by Boss[99] in 1972.

In 1972 Volchok[100] et al. investigated the plutonium in the neighborhood of Rocky Flats as pertains to airborne respirable particles. Experiments were carried out to determine the respirable fraction of the resuspended ^{239}Pu. The results indicate that in this area, with concentration in excess of 1 fCi/m^3, the respirable fraction is about 0.2 to 0.4.

In 1972 Fish[101] et al. discussed experimental data and analytical models to be considered in assessing the transport of plutonium aerosols following a hypothetical reactor accident. Behavior of released airborne materials within the reactor containment system, as well as in the atmosphere near the reactor site boundaries, were semiquantitatively predicted from experimental data and analytical models.

The results of a simulated airborne release of uranium, representing plutonium, during the burning of contaminated waste were discussed by Mishima and Schwendiman[102] in 1973. A source term containing both a fractional release and aerodynamic size distribution is required to evaluate the potential downwind hazard from a postulated inadvertent release. The approach taken in this study of a potential accident situation in a radiochemical processing plant was to measure the fractional airborne release and aerodynamic size distribution released using a simple but realistic situation. The release of uranium (as a stand-in for plutonium) was measured during the burning of flammable, solid materials containing either uranium dioxide powder or uranium nitrate solution. The flammable materials used were representative of the typical types of solid waste generated by such a process and were packaged in a typical manner.

In 1972 Mishima and Schwendiman[103] characterized radioactive particles in a plutonium processing plant exhaust system. Filter and cascade impactor samples were taken of the stack gases and various exhaust streams. The aerodynamic characteristics, the amount and distribution of particles and their associated radioactivity were measured. The general conclusion that was reached was that the plutonium present appeared to be attached to large, nonactive particles.

A series of studies to determine the fraction of plutonium made airborne and characteristics of aerosols produced by overheating plutonium metal and several plutonium compounds was undertaken by Schwendiman[104] et al. in 1958. Plutonium in various phases of processing and use commonly appears in three forms--plutonium metal, plutonium compounds as powders, and liquids containing soluble plutonium compounds. All three forms can be found in various atmospheres and configurations. Of the materials studied, plutonium compounds in the form of powders released the largest amounts of plutonium aerosol. The chemical compound, temperature and airstream velocity obviously had a great effect on the percentage released. The highest rate of release was from partially oxidized plutonium oxalate, 0.82 wt % per hour at 1000° C in 100 cfm per second air. Plutonium fluoride release rates were the lowest for the compounds considered, 0.007 to 0.05 wt % per hour.

Overheating plutonium metal created less airborne material than its compounds. The size distribution of the oxide produced can vary with the oxidation conditions and, since the oxide is friable, the mechanical processes to which it is subjected. The median mass diameter of the particles airborne during one of the experiments was 4.2 micrometers.

Heating liquids containing soluble plutonium compounds released the smallest amounts of material. Using slow heating rates, the maximum amount airborne from a concentrated plutonium nitrate solution over a 2-hour period was 0.03 wt %. When a greater heating rate and greater volume were used, the maximum amount airborne was 0.18 wt % during a 63-minute sampling period at a full rolling boil. Tables are presented in the report giving the results of the study in greater detail.

In 1973 Clarke[105] devised a model for determining dispersion of effluent from stacks of nuclear installations and evaluated the inhalation doses.

Plutonium isotopes ^{239}Pu, ^{240}Pu and ^{238}Pu have been injected into the stratosphere as a result of atmospheric nuclear weapons tests and have reached the ground as particulate fallout. A satellite bearing a Systems for Nuclear Auxilliary Power generator (SNAP-9A), containing 17 kCi or 1 kg of ^{238}Pu reentered the atmosphere in the southern hemisphere in 1969. The resulting burn-up during reentry turned the ^{238}Pu into small particles at an altitude of about 50 km. The global inventory and distribution of fallout plutonium from these and other sources were compiled by Hardy[106] et al. in 1973. The surface air concentrations and characteristics of the SNAP-9A burnup in the vicinity of Richland, Washington, were also reported in 1973 by Thomas[107].

A mathematical model describing the behavior of particulate plutonium in the lower atmosphere was proposed by Travis[108] in 1973. The model accounts for particles settling or impinging on the earth's surface as well as particle re-entrainment or resuspension.

The radioactive fallout rates and mechanisms describing the behavior and characteristics of radioactive debris from Chinese nuclear weapons tests were reported in 1973 by Thomas[109] et al.

The nature and distribution of radiation to be expected in the stratosphere, including that resulting from nuclear tests, contributions from plutonium released on burnings of SNAP generators, galactic rays and solar proton flares were included in a report by Webb[110].

Water

The dissolution of ^{238}Pu in environmental systems was the subject of a report by Patterson[91] et al. He points out that some experimenters have erroneously assumed that the problem of plutonium transport is connected with the "solubility" of PuO_2, whereas the real problem is the rate of dissolution. Plutonium dioxide reacts chemically with aqueous solution to release plutonium ions. These ions, in turn, will react quickly with other agents in the solution to form soluble complexes or will react with the water itself to form insoluble suspensions. The term "solubility" does not apply in the thermodynamic sense to the plutonium dioxide water system. Because the dioxide cannot be formed in the presence of water, no equilibrium can be achieved between the dioxide and the plutonium ions in solution. The dissolution reaction for PuO_2 is irreversible, the dioxide continuously goes into solution. In a static system a state of pseudo-equilibrium may be reached, with the plutonium in solution remaining constant. The concentration of ionic species of plutonium is controlled by the solubility product of the hydrous oxides that are precipitated from solution according to Polzer[111] in 1971. The rate of dissolution of the dioxide is independent of the concentration of the plutonium ions in solution.

As stated in 1974 by Patterson[91] et al., the definition of plutonium in solution is not well defined. The "solution" may contain fine particles of undissolved oxide, precipitated hydrous oxide, polymers in colloidal form, and various complex ions of plutonium.

As shown by Polzer[111] and Silver[112] in 1971, the rate of dissolution of PuO_2 is dependent on many factors, including pH, temperature, oxidizing, reducing, or complexing agents and surface areas of the oxide (particle size and shape).

The history of the sample is also important, including the duration and temperature of heat treatment, time, temperature, and radiation history since treatment. According to Kapshukov[113] et al. in 1971, the high specific alpha activity affects several of these factors, $^{238}PuO_2$ might be expected to dissolve more rapidly than $^{239}PuO_2$. The self-irradiation of the $^{238}PuO_2$ causes damage to the crystal lattice, but this damage may be annealed out by heat treatment. It was found that the lower the temperature at storage, the greater the retention of damage.

In 1974 Patterson[91] et al. concluded that PuO_2 initially rapidly dissolves or reacts when first placed in contact with a neutral aqueous medium. The rate may vary from sample to sample under nominally the same conditions but is generally greater than 100 ng/m^2s for ^{238}Pu and on the order of magnitude of 1 ng/m^2s for ^{239}Pu. After a few hours the rate decreases to a value that generally remains constant, until the concentration of alpha activity becomes high enough to initiate competing reactions, such as coagulation of suspended material or precipitation from a colloid. The change in rate of dissolution after a few hours is unknown but in 1969 Kubose[114] et al. speculated that a hydrous coating may form, protecting the material from further rapid dissolution.

In an electrochromatographic experiment, Lingren[115] discovered in 1968 that the dissolution of high-fired PuO_2 in seawater yielded negatively and positively charged carbonate complex ions as well as neutral colloids. The latter may be in the form of very small hydrous oxides as stated by Lloyd and Haire[116] in 1973.

The dissolution of airborne plutonium in water has been measured in a scrubber in parallel with an air filter by Hayden[117] in 1972.

Some investigations have been done on the dissolution of $^{238}PuO_2$ microspheres in water (Hudson[118] in 1968; Adams and Fowler[119] in 1974). In 1973 Raabe[120] et al. reported on in vitro solubility of respirable particles of ^{238}Pu and ^{239}Pu.

A report by Curtis and Bentz[121] in 1972 discusses the chemistry of plutonium in natural waters. The assumption that the form of plutonium in natural water would be that of the tetravalent state is questioned. Accidentally released "soluble" plutonium is alleged to form plutonium-polymers, or becomes solubilized ionic trivalent plutonium.

Nature provides an abundance of organic materials which may act as reducing agents toward an oxidizing material such as the Pu + 4 ion, so that were the Pu(IV) polymer to depolymerize, it might yield trivalent plutonium ions. On the other hand, plutonium dioxide falling into ocean waters may spend years dissolving. As radioactive decay and natural causes slowly degrade the oxide, plutonium may be released to the ocean water very slowly and at such low concentrations that polymer formation may not be possible. Several things may occur to the released soluble plutonium: it may become attached to a bit of sediment or organic material and remain so attached in the tetravalent state; it may be reduced to Pu(III); or it may be oxidized to a higher form. A computer program for predicting the final form of "soluble" plutonium in given conditions of acidity, potential, and complexation factors was proposed by Silver[122] in 1971.

In 1972 Dix and Dobry[123] summarized some of the environmental phenomena that modify plutonium reactions in water. They listed the results of Metz who placed plutonium sulfate solution in synthetic seawater and found that it "disappeared" from solution with a half-life of about 40 hours. Of the plutonium recovered, 50% was found in a tan flocculent deposit on the bottom of the tank, 20% was found on the walls of the tank below the surface and about 30% was found on the walls above the water line which was exposed to the spray from air bubbles resulting from circulation and aeration. Metz's experiments with a fresh specimen of $^{238}PuO_2$ molybdenum cermet yielded surface dissolution rates of 0.01 μCi/day per square millimeter of exposed surface area (0.05 μg/day per g of PuO_2) and plate out of an insoluble plutonium product was observed on the container surface.

In 1972 Langham[124] studied the fate of PuO_2 following the Thule accident. He found that particles agglomerated on inactive debris and only about 1% was suspended as very fine particulates in water derived from melting ice. In 1966 Lai and Goya[125] found that plutonium metal chips placed in seawater reacted completely in about 10 hours, accompanied by the evolution of hydrogen which served to continuously expose fresh metal surfaces until the reaction went to completion. The products of the reaction were small particles of black material (PuO_2) that settled to the bottom and a white or pale green gelatinous product [$Pu(OH)_4$] of low density which was suspended in the water. The solubility of the above products appeared quite low. As stated by Taube[126] in 1964 the equilibrium constant for hydrated plutonium oxide in distilled water is 7 X 10^{-56}.

In 1971 Aarkrog[127] re-examined the area of the accidental release of plutonium at Thule, Greenland. The investigation included samples of seawater, bottom sediments, bottom animals, zooplankton, fish, seabirds, seals, seaplants, and lichen. The results showed that the levels had decreased but plutonium was still present in concentrations significantly above the fallout background. Samples of bottom sediments showed a vertical movement of plutonium in the sediments down to a depth of at least 10 cm. Bivales as far as 30 kilometers from the point of impact contained plutonium from the accident.

In 1968 Kubose[128] et al. found that when $^{238}PuO_2$ microspheres, 100 μm in diameter, were implanted in situ in the Pacific Ocean on the surface of bottom sedimets they became encrusted as a result of biochemical action. The microspheres were clearly visible in each aggregate and appeared to be little altered by submergence after 5 months in the sea. The aggregates were composed of sand grains and organic detritus, which was lightly cemented around the microsphere by a dense matrix of mucilage and small diatoms. This phenomena lowered the rate of dissolution of $^{238}PuO_2$ markedly. Observations were also made on microspheres buried within bottom sediments. The plutonium adsorbed to the sediments and remained localized at the site of the microsphere.

The particulate size and physicochemical state of plutonium in the Bikini Atoll Lagoon is reported by Nevissi and Schell[129] in 1975. They found that 16 years following the last nuclear test on the Atoll, the plutonium was neither buried totally in the lagoon sediments nor has it been discharged completely to the ocean. Small particles and/or ions are released at the sediment-water interface and are transported by the currents. These particles and ions agglomerate during transport away from the sediment surface.

In 1972 an evaluation of the potential for concentration and transfer of plutonium in Lake Michigan was done by Wahgren and Nelson[130]. The present levels, distribution and biogeochemical behavior of fallout plutonium are presented.

BIOLOGICAL ASPECTS OF THE PHYSICOCHEMICAL COMPLEXITIES OF PLUTONIUM

A detailed summary of the chemical and physical properties of plutonium was given by Taylor[131] in 1973. Saenz and Ramos[1] also summarize the physical and chemical aspects of plutonium as applied to physiological interactions among biological systems. A brief summary of the most important biological aspects of the chemistry of plutonium in this review concerned its states of oxidation, its tendency to hydrolyze, and its capacity to form complexes. The polyvalence shown by plutonium from Pu(III) to Pu(VII), and the proximity of its oxidation potentials permit equilibrium states in solution among its ions of different valences. This coexistence brings about a very complex chemistry of this nuclide in its biological behavior.

Mammals

If plutonium in ionic form enters the mammalian body, three main types of reaction may be expected to occur.

Hydrolysis to yield polymeric or colloidal species - Plutonium can hydrolyze to insoluble hydroxide, passing through intermediate states of polymerization with formation of colloidal aggregates. This phenomena can occur at physiological pH, qualitatively and quantitatively influencing its distribution and deposit in the various systems, organs, and tissues.

Complexing by proteins or other biological macromolecules - Plutonium deposited in a localized area will complex with structural proteins. Particulate forms may be removed by phagocytic action.

Complexing or chelation with small molecular weight components of animal cells and tissues - Plutonium complexes with anions of mineral acids (nitrates, carbonates, etc.) and of organic acids (citrates, proteinates). Stability depends on the complexing bonders. Bonders of great chemical stability can form complexes which dissociate at physiological pH. Other nonbiodegradable bonders form biologically stable complexes. Thus, the interaction of plutonium with a broad variety of biological bonders produces simultaneous mechanisms of dissociation and formation of new complexes which influence retention, excretion and elimination therapy.

Plutonium compounds occurring as airborne contaminants are either very insoluble in body fluids, e.g., PuO_2, or are relatively soluble and transportable in the body, e.g., plutonium nitrate.

Insoluble plutonium translocates from the respiratory tract very slowly, over many years, and this imparts most of the emitted radiation energy to lungs and associated tissues such as thoracic lymph nodes. Insoluble plutonium inhaled as particles normally remains in the particulate form in the body for a relatively long time. The size of the particles determines the deposition and mobilization in the respiratory tract. The transportable compounds inhaled are diffused in the blood and organic fluids, and are transported to be deposited in organic tissues or excreted in the urine.

Of the nontransportable compounds, a fraction of that deposited in the upper respiratory channels is exhaled and another part is moved by ciliary action toward the esophagus and gastrointestinal tract. The remaining fraction which is deposited in the lower portion of the lungs is either localized or is slowly moved either toward the gastrointestinal tract and eliminated in the feces, or toward the blood and lymph by solubilization of the particles or by transport through the pulmonary epithelium. The mechanics of elimination of radioactive particles from the respiratory tract are basically ciliary action, phagocytoses and transport to the circulatory system (blood and lymph).

Saenz and Ramos[1] presented the kinetics of movement of plutonium particles in the respiratory tract and Bair[132] et al. in 1973 summarized the deposition and translocation of inhaled plutonium in great detail.

Diffusion of plutonium in the blood and interaction with proteins are discussed by Saenz and Ramos[1] and Taylor[131]. Plutonium reaching the blood may be absorbed and excreted in the urine without change, if the compound is stable; disappear in a few minutes to be mobilized by the liver, if strongly hydrolyzed; or form complexes with proteins if the compound is unstable. Approximately 90% of serum protein is protein bound, mainly to the beta globulin (transferrin) to form a very stable complex (Palmer[133] in 1956 and Belliayer[134] in 1959). The interactions of plutonium with protein are influenced by the physicochemical characteristics of the plutonium compound that reaches the blood.

Many authors have studied the interactions of Pu(IV) and serum proteins to establish that transferrin is the major plutonium binding protein in man [(Stover[135] et al. in 1968; Bruenger[136] et al. in 1970); dog (Stevens[137] et al. in 1968; Muntz and Barron[138] in 1947); horse (Turner and Taylor[139] in 1968); rabbit (Taylor[140] in 1969); and rat (Boocock and Popplewell[141] in 1965 and Turner and Taylor[142])].

As summarized by Taylor[131], transferrin is a glycoprotein with a molecular weight of about 90,000 in which aspartic acid, glutamic acid, lysine, leucine and alanine are the most abundant amino acids. The transferrins of all the species studied bind 2-gram atoms of iron to form a complex which dissociates in acid solution but is stable up to pH 9 to 10 as reported by Feenez and Komatsu[143] in 1966. The effect of pH on the binding of Pu(IV) to transferrin was studied by Chipperfield and Taylor[144] in 1970, who showed that maximum binding occurred at pH 7. The mechanisms of Pu(IV) binding by transferrin are not fully understood. In 1968 Stover[135] et al. and Turner and Taylor[139], on studying dog and horse sera, concluded the bicarbonate ions were required for the binding of Pu(IV) to transferrin in a similar manner to their requirement in the binding of iron (Feenez and Komatsu[143]).

Chipperfield and Taylor[144] question the requirement for bicarbonate in the binding of Pu(IV) to transferrin. Plutonium can be displaced from its combination with transferrin by the addition of iron; little plutonium is found bound to iron-saturated transferrin (Popplewell and Boocock[145] in 1968; Stevens[137] et al.; Turner and Taylor[139]; Massey and Lafuma[146] in 1968). Under their experimental conditions, Chipperfield and Taylor[144] found that the binding of iron and plutonium to transferrin was similar in that both increased up to pH 7; above pH 7 the Pu(IV) transferrin complex was less stable than the Fe(III) complex, but this may be a reflection of the greater tendency of plutonium to hydrolyze. The results of these studies indicate that Pu(IV) can bind at the iron binding sites in transferrin but can also bind at other sites in the molecule.

The stability of the Pu(IV) transferrin complex has not been determined. Stover[135] et al. concluded it had a high stability but less so than Fe(III) transferrin. Popplewell and Boocock[145] showed that Pu(IV) can be displaced by citrate or DTPA.

A small proportion of plutonium may be associated with albumin and with the gamma and other globulins (Turner and Taylor[139]; Stevens[137] et al.; and Popplewell and Boocock[145]).

As summarized by Saenz and Ramos[1] the distribution from the blood and deposit in the various organs and tissues are determined by the stability of the plutonium compounds which have passed to the blood or which have been formed by hydrolysis at physiological pH. In the case of heavily complexed compounds, deposit takes place preferentially in the bone at a proportion of about 80% to that in liver. If the compound has been hydrolyzed, the deposit takes place preferentially in the liver, at a proportion of 70 to 80% to that in bone.

Deposit of plutonium takes place on the surface of the bone as found by Stover and Eyring[147] in 1970, mainly in the cells of the endostium, and to a lesser degree on the periostium. The selectivity for the cells of the endostium is associated with the presence in those cells of a glycoprotein, sialoprotein, to whose carboxylic groups plutonium is joined to form a very stable complex (Chipperfield and Taylor[144] and Herring[148] et al. in 1962). This incorporation includes mechanisms for the dissociation of the Pu-transferrin complex at the level of the bone surface and diffusion of the plutonium from the blood, with the changes of pH derived from the presence of specific organic acids of the bone (lactic, citric) mainly influencing this process.

Experiments with different species of animals and in different parts of the skeleton in 1972 showed the nonhomogeneity in the distribution in the bone in medullary spaces, and later, according to Jee[149], at the base of the cranium.

Plutonium can be localized in the red bone marrow before being deposited in the bone medullary spaces, and later, according to Jee[149] and Erleksova[150] in 1960, the plutonium mobilized from the bone deposits can be found either recirculating in the blood, or in spaces in the bone marrow, or retained in cells of the reticuloendothelial system.

The accumulation and retention in the medullary macrophages seem to be associated with processes of phagocytosis and existence of the iron storage protein, hemosiderin (Vaughn[151] et al.). Their conclusion is that plutonium reaching the blood by any route of entry, if in monomeric form, is deposited initially in the skeleton rather than in the liver; if in the polymeric form, it is likely to be deposited in the liver rather than the skeleton though it may subsequently translocate to the skeleton. During their research of the subject they concluded that little is known at present about the mechanisms and the form in which plutonium translocates from the site of entry to the blood stream, though experimental results suggest it is initially translocated in the form of soluble complexes and may later take a more colloidal form. Initially it can be expected to be deposited primarily in the skeleton rather than in the liver. Plutonium, from occupational and fallout exposure, reaching the circulation in man after some years, has been assessed as redepositing roughly half in bone and half in liver (Taylor[152]).

The fractions of available plutonium which are deposited in the liver and the characteristics of their distribution are a function of the physicochemical state of the compound and of the route of penetration (Saenz and Ramos[1]). In general, low relations of liver/bone deposits indicate the entrance in blood of complexed transportable compounds. High values suggest the existence of hydrolized plutonium.

By intravenous injection of complexed compounds in dogs and rabbits, Cochran[153] et al. observed in 1962 a diffuse initial distribution in all liver structures and, later, an irregular redistribution and accumulation in cells of the endothelial network according to the degree of polymerization reached in their interaction with organic fluids.

With easily hydrolyzable compounds, the initial distribution is not homogeneous, and the plutonium aggregates are localized in macrophages mainly in peripheral zones of the hepatic lobes (Lindenbaum[154] et al. in 1957).

Mechanisms of interchange between plutonium and proteins seem to be involved in incorporation into the liver, especially processes of phagocytosis in redistribution.

In this respect, histological studies of dog livers in vivo (Stover and Eyring[147] in 1970) and in vitro (Bruenger[136]) show the existence of plutonium associated with ferritin in the liver. This leads to the supposition that the incorporation takes place according to the following reaction:

Pu IV-transferrin + ferritin = transferrin + Pu IV-ferritin.

The processes of phagocytosis, on the other hand, facilitate the mobilization and interchange of plutonium between the hepatic compartment and the Kupffer cells.

Plants

In the area of plant uptake of plutonium, the determining factors are plant species and vigor, soil type and chemical form or solubility of the deposited material (Romney and Davis[155] in 1972 and Menzel[156] in 1965).

Plutonium is tenaciously held by the soil, preventing plant incorporation, particularly if salts, acids, detergents and organic compounds are absent (LASL[37] in 1974). Soil characteristics, such as soil structure, organic content, pH and the amount of clay present, influence the availability of plutonium to some extent but probably no more than a factor of ten (Romney and Davis[155]).

BIOLOGICAL DEPOSITION AND EFFECTS

DEPOSITION AND EFFECTS OF PU IN AQUATIC ENVIRONMENTS

When plutonium is released to surface or groundwaters it may be concentrated by and in plants and in silt sediments and suspended materials in the water. The concentration of plutonium in sediments or in aquatic organisms frequently exceeds their concentration in the surrounding water, often by several orders of magnitude as reported by Sayre[157] et al. in 1963. The plutonium, thus concentrated, may be released suddenly at some future time in quantities far exceeding their maximum permissible concentrations.

The available data concerning the dissemination of plutonium in the aquatic environment were summarized by Noshkin[158] in 1971. The most studied isotope has been ^{239}Pu derived from worldwide fallout. Essentially all the published work has been concerned with levels in the marine environment where plutonium is found widespread among planktonic, pelagic and benthic organisms. The concentrations are higher in organisms feeding on sediment or on surfaces than in those drawing on the water itself. There is some evidence that plutonium concentrations are increased in organisms of high trophic levels. Bone and liver are major repositories for plutonium in marine vertebrates while muscle tissue of both marine vertebrates and invertebrates contain relatively lower concentrations. In marine sediments, as in soils, plutonium is more mobile than was originally expected. Fallout ^{239}Pu appears to contribute more than fallout ^{90}Sr or ^{137}Cs to the artificial radiation exposure of many marine species.

The results of studies of the concentration of plutonium in marine invertebrates of the North Atlantic Ocean and their ecological relationships are presented by Noshkin[159] et al. Organisms from the near-shore environment were selected to show effects on plutonium uptake of variations in feeding habits, association with sediment, or with absorptive surfaces and of trophic levels. Of the invertebrates analyzed, the mussel clam, oyster and scallops are filter feeders, subsisting on tiny organisms and organic detritus removed from suspension. Plutonium-239 concentrations in the mussel body average 51 dpm/100 kg, a level 300 times that in seawater. The shell of the mussel was found to have, on the average, 64% more plutonium than the same weight of soft tissue. Fallout plutonium concentration in the mussel has increased by a factor of 2 in 6 years, as compared to results of Pillar[160] et al., reported in 1964. Deposition as recorded in Tokyo by Miyake[161] et al. in 1970 has increased only by 25%.

The ^{239}Pu concentration in starfish collected as they were feeding on the mussel beds, although different in each sample, was about four times that found in the mussel upon which each was feeding (Noshkin[159]). The marine worm, *Nereis*, contained the highest concentration found in marine invertebrates. This worm is a nonselective deposit feeder and ingests quantities of surface sediment. Concentrations in other invertebrates are also given in the report by Noshkin[159] et al. Significant concentrations of ^{239}Pu were also found in Sargasso weed. This weed may cycle considerable quantities of ^{239}Pu to specific near-shore regions.

In 1971 Wong[162] et al. reported on the plutonium concentrations in organisms of the Atlantic Ocean. The results showed that ^{239}Pu was concentrated in some tissues of each organism so far examined. In fish, ^{239}Pu concentrations range from 0.2 to 140 dpm per 100 kg fresh weight; in benthic invertebrates from 23 to 140 dpm per 100 kg; in plankton, from 130 to 340 dpm per kg and in Sargasso weed, up to 1280 dpm per 100 kg.

In 1971 Patin[163] et al. reported on a series of experiments performed to study the accumulation of ^{239}Pu by live and dead *Misgurnus Fossilis* spawn. The intensity of the accumulation was found to be related to the phase of embryogenesis because of change in the membranes of the spawn. The bulk of the plutonium was adsorbed on the membrane of live spawn. In dead spawn, the adsorption of plutonium was virtually irreversible.

Wong[164] et al. in 1970 reviewed data from a number of studies of ^{239}Pu uptake and retention by marine animals and sediments and the content of ^{239}Pu in seawater from various locations. It was concluded that most of the plutonium released to the near-shore marine environment concentrates in sand and silts of the sea floor and beaches.

Hodge[165] et al. in 1973 found that repeated measurements of ^{239}Pu in several organs of albacore tuna suggest that the upper layers of the North Pacific Ocean can retain large fractions of this nuclide for periods of a decade or more. During the period from 1965 to 1971 the reported rate of input from fallout decreased by about one-half in 1 year. During this same period, ^{239}Pu concentrations in albacore liver decreased to one-half in about 3.5 years.

In 1974 Adams and Fowler[166] placed goldfish in aquaria containing $^{238}PuO_2$ microspheres. At intervals up to 302 days the fish were analyzed and tables were presented to show alpha activity in the gills, intestine, muscles, bones, and total body. Results are also presented for radioactivity in the aquaria water and for radioactivity of the shell and tissues of inhabitants of the aquaria after 185 days.

Examples of variations in bioconcentration and concentration factors for plutonium in aquatic ecosystems are given by Kneip and Lauer[167] in 1973. Problems associated with making general statements regarding concentration factors are discussed.

Two reports covering the years 1971-72 and 1973 present the data of studies on the plutonium concentration along freshwater food chains of the Great Lakes (Ouchi[168] et al. and Bowen and Noshkin[169]). Results indicate that bottom feeding fish, e.g., shad, sucker, carp, bullhead, and rum, had higher ^{239}Pu concentrations than predators such as perch or bass.

Results of studies to determine the ^{239}Pu levels in water, sediment, plankton and fish samples collected from Lake Michigan were reported by Marshall[170] et al. in 1972.

Gromov and Spitsyn[171] in 1974 studied the change in the physicochemical state of ^{239}Pu due to its assimulation by phytoplankton. The stabilization of the physicochemical form of ^{239}Pu alters the state of plutonium in the marine environment, which, in turn, leads to variation in the entrapment of this element by bottom sediment and by suspended matter.

Gromov and Spitsyn[172] also studied the absorption of ^{239}Pu by a culture of single-cell green algae and by a natural phytoplankton community. It was found that the natural community concentrated the plutonium to a greater extent than the pure culture did. At 15 to 17 days, the isotopes are released into the seawater in a physicochemical form in equilibrium with the oceanic environment.

Folsom[173] et al. presented data in 1972 on the concentration of ^{238}Pu, ^{239}Pu, ^{240}Pu in IAEA seaweed samples.

In 1971 Wong[174] et al. found that seaweeds concentrate plutonium and that seaweeds may be a sensitive indicator for the detection of variations of plutonium concentration in the marine environment.

Zlobin and Perlyuk[175,176] in 1973, studied the photosynthesis and mechanism of the action of cyanide on cell respiration and ^{239}Pu accumulation by marine algae. The introduction of this respiratory inhibitor into the energy system of algae caused a response reaction in the form of a decrease in plutonium accumulation by plant cells. It is postulated that assimilation of colloidal particles of plutonium occurs as a result of ultraphagocytosis.

A study of plutonium and polonium inside giant brown algae is reported by Wong[177] et al. in 1972. It was found that large variations occurred between specific tissue types. Sampling thin parts of brown algae or thin outer layers should provide great sensitivity for detecting early changes in the extremely small traces of plutonium that are now anticipated in the sea from reactor effluents, nuclear fuel processing or fallout.

DEPOSITION, EFFECTS AND COUNTERMEASURES OF PU IN SOIL, PLANT ENVIRONMENTS

Soil

A great deal of study has been done concerning the deposition, concentration and mobility of plutonium in the soil of the Nevada Test Site (NTS). Langham[178] et al. in 1966 and Langham[179] in 1968 assessed the problem of plutonium contamination at NTS and elsewhere and suggested that the hazard to man was minimal as long as particles deposited on vegetation and soil remain deposited. In 1970 Romney[180] et al. observed a slight downward movement into undisturbed soil profile at NTS during the 10 years following deposition. Although the solubility of plutonium fallout particles in the presence of soil is poorly understood, plutonium infiltration to a depth of 12 cm was attributed to the downward movement of high density particles and not solubilization. In 1970 Mork[181] noted very similar downward movement in the same sampling area 2 years post-detonation where plutonium was found to be predominantly associated with >44 μ particles. Price[182] in 1973 reviewed the distribution and fate of plutonium in terrestrial ecosystems and identified areas needing further study.

In 1973 Tamura[183] et al., discussed studies characterizing plutonium-soil interactions at NTS. Samples were taken to provide adequate activity levels to permit determination of plutonium on different size and mineral fractions. The soils were then subjected to extraction of the plutonium content by nitric and hydrofluoric acid.

The ecological aspects of plutonium dissemination in terrestrial environments at the NTS were covered in a report by Romney and Davis[155] in 1972. Emphasis was placed upon standardization of analytical methods, delineation of contaminated areas, problems of resuspension and redistribution, food chain transport and ecological effects.

Anspaugh[184] et al., Eberhardt[185] and Gilbert[186] gave summaries of test results and statistical analysis of soil plutonium studies at NTS. Inventory of ^{239}Pu and ^{240}Pu at various depths in surface soil samples was determined and results of resuspension and redistribution studies were given.

Soils of five areas located on the Test Range Complex at NTS were characterized and soil profiles prepared, identified, and classified by Leavitt[187] in 1974.

The plutonium content of soil at Nagasaki, Japan, 24 years after detonation of a nuclear device, was reported by Sakanoue and Tsuji[188] in 1971. The results indicate that plutonium contaminants in soil are not easily removed by natural forces.

The literature regarding effects of the liquid waste disposal operations at Hanford on the soil environment was summarized by Rautson[189] in 1973. The summary is divided into studies pertaining to: the vadose zone below a depth of 6.1 meters (lower vadose zone); the saturated zone; and the vadose zone above 6.1 meters (upper vadose zone). Migration of wastes discharged to the lower vadose zone over the past 20 years by the mechanisms of diffusion, leaching and particulate transport is discussed.

A program to examine soil-actinide relationships in sediments from a disposal facility and characterization of actinide-bearing soils was covered in a report by Ames[190] in 1974. At least two types of plutonium were found by autoradiographic and microprobe examination of core samples. Plutonium particles (up to 10 μm in diameter and 60 wt % PuO_2) were the most conspicuous form. The second form of plutonium occurred in lesser concentration (<0.4 wt % PuO_2) but was found associated with silicate hydrolysis.

The isotopic analysis of soils in the vicinity of the Lawrence Livermore Laboratory as measured in 1973 was given by Silver[191] et al. in 1974.

Mass isotopic analysis of fallout in the soils of north central and southeastern sections of Utah collected in 1971 indicates that the NTS was the probable secondary source according to a report by Hardy[192] et al. in 1972. Isotopic ratios and composition of the fallout in soil were given.

Environmental monitoring reports by Reynolds Electrical and Engineering Company, Inc.,[193] in 1969 and the National Environmental Research Center[194] in 1973 gave the soil fallout composition from testing activity as determined in areas surrounding the NTS. Plutonium-239 soil measurements, depth profiles and physical state were given for the Rocky Flats vicinity by Poet and Martell[86] in 1972.

In 1974 Healy[195] proposed an interim standard for the upper limit of concentration of plutonium in soils in inhabited areas. Available information on possible sources of exposure of people living in an area where soils are contaminated with plutonium was analyzed to derive estimates of intake. An evaluation of protective guidelines was reported by Anspaugh[184] et al.

Bliss and Dunn[196] reported in 1971 on the results of studies underway to determine plutonium in soil from areas outside the NTS. Plutonium was detected in four locations and showed concentrations in the top 3 cm of soil that were 10 to 100 times greater than the concentration of soils in other areas.

The role of soil microorganisms in the movement of plutonium was noted by Au[197] in 1974. The study was designed to determine the ability of microorganisms to absorb plutonium, to quantify the uptake and to determine the microbial population of soils of the NTS.

The effects of soil heating by ^{239}Pu on field flora were studied by Krivolutskii and Fedorova[198] in 1973. The results indicate a sharp decrease in the number of soil organisms as a result of heating the medium with ^{239}Pu.

The feasibility and environmental impact of cleaning up plutonium contaminated areas in Nevada were discussed by Wallace and Romney[199] in 1974. It was felt correction of the damage done to the fragile environment during the course of decontamination may require greater efforts than the decontamination. Alternate procedures were discussed.

Plants

In 1973 Francis[200] found that a survey of literature associated with the movement of plutonium in soil and uptake in plants reveals that a major portion of the investigations pertains to soils developed under arid or semiarid climates.

Short-term studies summarized by Price[182] in 1973 indicate less than 10% plutonium uptake by plants. Fallout plutonium uptake by plants was studied by Nishita[201] et al. in 1965 who reported that ladino clover uptake was very low. Another study by Selders[202] et al. in 1955 found that only trace amounts of alpha activity were detectable in tomato plants grown in fallout-contaminated soil, and no uptake was recorded for bean, barley or tumbleweed. In 1948 Jacobson and Overstreet[203] reported that plutonium uptake by barley from clay suspensions was greatest for tetravalent plutonium. That the sorption of tetravalent plutonium to root surfaces from culture solutions was linear with respect to concentrations, whereas, leaf concentrations appeared to be curvilinear, was also noted in 1955 by Rediske[240] et al. The uptake into shoot tissue by tumbleweed from solution cultures was slightly less than for bean, barley or tomato.

In 1971 Cummings and Bankert[205] used pot culture to compare the uptake by plants of ^{238}Pu from nine different soils. Plutonium uptake was found to be 7×10^{-6}% to 280×10^{-6}% of plutonium applied.

In 1963 Wilson and Cline[206] compared the uptake of plutonium from soil by barley with that of tungsten and lead. Using a modified Neubauer technique, it was found that uptake of Pu(IV) from an acid soil was greater than uptake from alkaline or calcareous soils, and plutonium uptake was less than tungsten or lead. Cline[207] in 1967 reported the uptake of americium and plutonium from two of the same soils and indicated concentration factor (CF) values [CF = μCi/g acceptor (e.g., plant)/μCi/g precursor (e.g., soil)] of 0.003 for americium uptake from either soil and 0.0002 and 0.0001 for plutonium uptake from acid and alkaline soil, respectively.

Neubold and Mercer[208] and Neubold[209] reported that although plutonium uptake by perennial ryegrass was low, it increased over the 2-year period of study. It was detectable in the third harvest of the first growing season for one of the three soils studied (an acid soil) and the fourth harvest showed barely detectable uptake from all three soils. Results from the next growing season indicated that uptake from the acid soil was nearly four times greater than the previous year's maximum. In 1970 Romney[210] et al. studied the uptake of fallout plutonium by clover over a long period under greenhouse conditions. A substantially greater foliage uptake of plutonium, a sevenfold increase, from a highly contaminated soil occurred after 5 years of cropping as compared to the first year. It was postulated that the increase in uptake was due to either more perennial roots coming into contact with plutonium particles as the plants aged or that natural organic materials resulting from root tissue decay complexed with the plutonium and resulted in increased uptake.

As chelates enhance plant uptake of heavy metals, the influence of chelating agents on the uptake of plutonium was studied by Hale and Wallace[211] in 1970. They compared the effects of chelating agents DTPA, EDDHA and Fe-EDDHA on the uptake of heavy metals from soil by bush beans.

Romney[212] et al. reported results from studies of perennial vegetation growing in Area 13 of NTS. Relatively uniform distributions of $^{239-240}$Pu and ^{241}Am were found among individual samples of the same plant species. However, there were considerable variations in the contamination levels between different species, presumably from superficial entrapment of resuspended particulate material. Concentrations in *Eurotia lanata* were three to five times higher than in other species sampled from the same study site. Additional studies by Romney[213] et al. were reported in 1975.

Price[214] studied tumbleweed and cheatgrass uptake of plutonium and demonstrated that shoot uptake is clearly influenced by the chemical form of the transuranic. Test results indicate that organic acid complexes of plutonium such as oxalate or citrate can increase plant uptake when added to soil as compared to uptake from dilute nitric acid solutions.

In 1972 Smith[215] et al. studied the radionuclide content and botanical composition of the diet of beef animals grazing on the Area 18 range of the Nevada Test Site by analyzing rumen samples collected from fistulated steers. The radionuclide concentrations were generally low with periodic increases in individual isotope levels which could be traced to a specific contaminating event.

Klepper and Craig[216] in 1973 report studies of the interactions of plutonium aerosols with plant foliage. The studies include exposure of 2-week-old bean seedlings to ^{239}Pu nitrate aerosols of 1 to 3 μ size at wind speeds of 16.4 m/min.

When samples of foliose and fruticose lichens were collected from 27 sampling sites in the Thule, Greenland, region following the crash of a bomber carrying nuclear weapons aboard, Hanson[26] found slightly higher than background amounts of ^{239}Pu and ^{240}Pu associated with particles of estimated 0.5 to 1.0 μm diameter.

In 1969 Alvarez-Ranies and Santos[217] studied the contamination of terrestrial gastropoda feeding on plants exposed to the plutonium aerosol from the collision of planes carrying atomic weapons over Palomares, Spain. Plutonium values in soil, plants and the gastropoda flesh and shell were tested and the interacting relationships noted.

ABSORPTION OF PLUTONIUM BY ANIMALS

Several possible routes of entry exist by which terrestrial animals can incorporate plutonium into their bodies. Herbivores and small animals may ingest radioactivity by foraging on contaminated vegetation, drinking contaminated water or consuming radioactive soil while eating or grooming the fur. Inhalation may also represent an important avenue of intake for burrowing animals and for animals living in an environment where considerable resuspension is evident. Predators, however, would probably derive most of their intake from the consumption of smaller prey animals which contain plutonium in their internal organs or on their pelts. Man enters into this terrestrial food web by consuming the meat from certain sport game and domestic grazing animals and the milk from dairy herds (LASL[37]). Ingestion has been considered to be the primary route of entry involved in the food chain transport of residual plutonium in the environment (Olafsen and Larson[218]).

Ershov[219,220] et al. in 1970 and 1971 studied the penetration of ^{239}Pu through the epidermis during superficial contamination using piglets. The results, shown graphically, indicate that at the beginning, activity penetrating the skin at any fixed depths very rapidly increased. After 12 hours of contact the amount taken up per unit time decreased and became balanced, i.e., the amount of the absorbed and expelled radioactive preparation was equal. The kinetics of microdistribution in the skin and the dose distributions in structural layers of skin are also given.

An early work by Finkle[221] in 1945 considered the distribution of plutonium in a dog 16 days after administration of a lethal dose of plutonium nitrate. The skeleton contained 25% of the injected dose, liver 31%, muscle 8% and spleen 3.5%; 10% was excreted. The plasma fraction of the blood contained 80% of the plutonium found therein. Studies with intravenous injection of plutonyl nitrate into mice found concentration of 27% of the dose after 64 days. When plutonyl citrate was injected the liver content fell from 35% to 14% on the 31st day and to 7% on the 64th day showing a half-time value of 20 days. Mice receiving plutonyl nitrate intramuscularly had liver retention that showed the same 20-day half-time. Plutonyl citrate injected into the peritoneal cavity of mice was slowly absorbed with a concentration occurring in the femur and liver; plutonyl nitrate, intraperitoneally, behaved similarly but was somewhat more slowly absorbed and therefore, was retained longer by the liver.

In 1948 Hamilton[222] noted that plutonium is not absorbed to any significant degree by way of the digestive tract. The metabolism of plutonium following intramuscular injection is essentially the same after administration as Pu^{+3}, Pu^{+4} and Pu^{+6}. He suggests that plutonium is converted by the body into one valence state regardless of the valence administered.

Results of the biomedical program in conjunction with the 1957 "Operation Plumbbob," were reported by Stannard[13] in 1973. The primary purpose of the program was the direct determination of the uptake and retention of plutonium by a variety of animals as a function of time of exposure and the relation of the amounts found to air and ground levels. There were two major phases and groups of exposed animals--those exposed during cloud passage and those exposed during long-term post-event periods. Rats and dogs were used in the former; dogs, sheep and burros for the latter. For the cloud passage phase, animals were placed from 164 to 656 meters downwind from Ground Zero. For the resuspension phase the animals were placed in areas with levels of ground contamination of 2.6, 40 and 560 micrograms per square meter of desert floor soil. The animals remained in place for periods ranging from 4 to 160 days. The plan was to measure the accumulation of plutonium in the lung and other tissues as a function of time and various aerosol parameters.

The dogs exposed to the cloud passage at the time of detonation showed generally higher burdens than the animals exposed to resuspended plutonium, even though the former were not at the range of maximum airborne concentrations at ground level. There was no appreciable increase in lung burden with time and very little difference in burden in animals placed at several different surface contamination areas. This may have been difficult to ascertain as radiochemical analysis at that time lacked sensitivity for very low levels.

Results reported from Operation Roller Coaster have not all been declassified at this time but what is available is included in papers by Wilson and Terry[16,17] and Stewart[223] et al. The initial lung concentrations for dogs, sheep and burros were very similar despite wide differences in lung size, etc. There was, however, a marked difference in clearance pattern among the three species. The lung clearance for the dog closely resembles that found in laboratory studies with inhaled PuO_2.

A review of the comparative metabolism of radionuclides in mammals was published by Stara[224] et al. in 1971. They note that the rate of absorption through the gastrointestinal tract was low when ^{239}Pu was given to rats. Katz and Weeks[225] reported absorption was 0.001-0.004%. Plutonium-239 $(NO_3)_4$ orally administered to rats was found to be absorbed between 0.002 and 0.003%, with a maximum of 0.009%. Carritt[226] et al., demonstrated in 1947, that the amount absorbed is related to the dose; with a lower dose, the absorption was 0.30%, whereas with a dose approximately tenfold higher, the absorption was decreased to 0.01%. Absorption of $^{239}Pu\ (NO_3)_4$ given orally to swine was reported as 0.002% by Weeks[227] et al. in 1956. Inhaled $^{239}PuO_2$ in dogs was absorbed in a range from 0.1 to 16.9% with a mean value of 3.7% as calculated by Morrow[228] et al. in 1967.

In 1972 Kashima[229] et al. conducted studies to clarify the relation between the physicochemical form of plutonium and its distribution in all organs and tissues after subcutaneous injection in mice. The forms used were monomeric plutonium with pH adjusted to 1, 4.5, 7.2, and 13.0 immediately before injection; and polymeric plutonium solution with its pH adjusted to 9.0. After subcutaneous injection of the monomeric form, ^{239}Pu remained mostly at the injected site for the 56 days studied. Concentration of plutonium in bone increased with time, the level of plutonium in liver, especially around the central vein, was relatively high in the initial 3 days, but was lower at 21 or 56 days. The distribution pattern was only slightly affected by changing pH or time prior to injection after preparation. With polymeric plutonium, however, relatively low plutonium concentrations were observed only in the bone and liver.

Ballou[230] et al. in 1972 found that about 0.08% of an oral dose of ^{239}Pu citrate was retained in dogs after 3 days, distributed mainly between skeleton (50%) and liver (25%). After intravenous administration the liver retained 32 to 45% of the dose and skeleton 29% of the dose 100 days post injection. Inhaled ^{239}Pu reached a peak concentration in blood after 6 hours, then decreased with a half-time similar to intravenously administered citrate. The lung, skeleton, liver, blood and intestines retained most of the inhaled ^{239}Pu 1 day after exposure. After 100 days, 36% of the initial deposit was retained; 33% in lung, 44% in skeleton, and 17% in liver. Plutonium was concentrated by lymphatic tissue following all methods of administration and all time intervals.

Morin[231] et al. administered ^{238}Pu or ^{239}Pu nitrate intramuscularly or by aerosol to 3-month-old rats. On the first day following intramuscular injection, almost no ^{239}Pu left the injection site; the amount of ^{238}Pu leaving the site of injection was higher. The first day following inhalation ^{239}Pu was cleared from the lung faster than ^{238}Pu. Blood and muscle uptakes of ^{239}Pu were 8 times as large as ^{238}Pu uptakes; ^{239}Pu activities in bone and liver were also higher. The metabolic fate of the isotopes was also different; there was greater early ^{238}Pu urinary excretion.

Baxter[232] et al. compared the total body distribution of monomeric and polymeric plutonium injected intravenously to beagle dogs and mice. Monomeric plutonium was cleared from dog blood less rapidly than polymeric plutonium. During the first 15 minutes only 20% (vs. 90%) had left the circulation. At sacrifice, 6 days post injection, significantly more monomeric plutonium remained in the blood although the amount of each form was less than 1%. Monomeric plutonium was deposited chiefly in liver and skeleton of both species. Polymeric plutonium was deposited in liver, spleen and lungs, with a comparatively small amount deposited in the skeleton. Autoradiographic measurements of plutonium deposition within the dog liver lobe showed a relatively homogeneous distribution of monomeric plutonium, whereas polymeric plutonium tended to be associated with sinusoidal (phagocytic) cells at the center of the liver lobe.

Taylor[233] et al. found that polymeric ^{239}Pu given intravenously to beagles was retained principally in the reticuloendothelial cells (RE) of the liver and spleen. Retention in the skeleton was limited to the RE cells of the bone marrow, with little, if any, uptake in the bone proper; except in the lung, where some mechanical filtration of larger particles may have occurred, the retention was principally governed by phagocytic factors.

Rosenthal[234] et al. in 1972 intravenously injected polymeric ^{239}Pu-nitrate (15% ultrafilterable) or of monomeric ^{239}Pu-citrate (90% ultrafilterable) into young adult female rabbits and sacrificed them at 3 days. The plutonium content of the intact femur (bone plus marrow) was higher in those receiving the polymeric form. Physical removal of the marrow from bone in the tibia showed the plutonium content of the bone to be slightly lower and of the marrow to be 17-fold higher after polymeric than after monomeric plutonium. Polymeric plutonium was also concentrated in spleen, ovaries, intestines (with contents), skeletal muscle and urine but less concentrated in liver. About the same levels of the two forms of plutonium were found in blood, bile and lung. For the first 30 minutes after injection, the polymeric plutonium was cleared from the circulation more rapidly than monomeric. After 60 minutes, the two forms were cleared at generally comparable, decreasing rates. Polymeric plutonium was as much as 10 to 30 times more concentrated in predominantly red (erythropoietic) marrow than in fatty marrow. Monomeric plutonium was 1.3 to 2.8 times more concentrated in the red marrow. Mice injected with the same plutonium solution as the rabbits showed, in comparison, a higher percent of the injected monomeric plutonium in the femurs and a lower percent in the liver; they showed a higher uptake of polymeric plutonium in the marrow and a higher uptake of both forms in the spleen and lungs than did the rabbits.

In 1974 Zapol'skaya[235] et al. intravenously injected male rats with ^{239}Pu chloride salt solution and determined the biological half-life in various tissues, organs and albumin proteins. The results indicated that the exchange of plutonium is determined by the exchange of the albumins with which they form bonds.

Durbin[236] et al. constructed a kinetic model to describe the transport and deposition of intravenously injected Pu(IV) citrate in the rat.

The implications of the solution of the model led to the following working hypothesis: (a) Unbound plutonium reacts with protein, presumably transferrin, in extracellular fluid compartments as well as in plasma. The Pu-transferrin complex is the most likely form in which orally administered plutonium is carried once it reaches the plasma. (b) Little, if any, protein bound plutonium is excreted or deposited in the liver. Formation of the Pu-transferrin complex is probably not necessary preliminary to liver deposition of diffusible plutonium. (c) Both unbound plutonium and bound plutonium are sources of plutonium deposited in bone. The surface of the reticulocyte (where iron is released from the Fe-transferrin complex) is considered the most likely site of dissociation of the Pu-transferrin complex.

A good review of the metabolism of monomeric and polymeric plutonium in small animals and the radiotoxic implications for man was prepared in 1971 by Lindenbaum[237].

Matsuoka[238] et al. compared autoradiograms obtained from animals administered monomeric or polymeric plutonium. Results were compared with those obtained from animals administered metabolically stable reference particles of known particle size. It was found that the behavior of plutonium soon after injection is largely influenced by its particulate character rather than by its chemical nature. Results following inhalation of a plutonium nitrate aerosol showed no obvious translocation to the liver, despite the water solubility of the nitrate form. The initial uniform distribution of inhaled plutonium changed to a nonuniform distribution after 3 months.

Baxter and Sullivan[239] studied the gastrointestinal absorption and retention of plutonium chelates administered to rats. They found the intestinal absorption of plutonium nitrate is increased about 700-fold when chelated with DTPA. Within 2 days, virtually all of the absorbed plutonium-DTPA complex is excreted in urine. Thus, plutonium retention in the liver and skeleton is quite low, but more than twice as high as when DTPA is not present. Citrate is less effective than DTPA in increasing absorption, but more effective in increasing retention.

In 1972 Mays and Dougherty[240] reported results of beagle studies. The initial distribution of plutonium was uniform throughout beagle liver following intravenous injection of monomeric plutonium in citrate solution. Most of the initial burden appeared in the hepatic cells.

Subsequently, much of the activity shifted into the liver reticuloendothelial cells lining the sinusoids; the rate of translocation was greatest at the higher dose levels, presumably due to release of the nuclide from cells killed by the irradiation. At very high doses, the skeletal burden may actually increase due to the amount released from the liver. At long times after injection the nuclide distribution is very nonuniform within the liver, being highest in the portal region and lowest in regenerative nodules that grow after the time of injection. In the lower-level ^{239}Pu beagles, the net liver burden remains roughly constant at about 30% of the injected activity for about the first 1,000 days after injection. The loss of liver ^{239}Pu during this period is at least, in part, compensated by the influx of ^{239}Pu released from the skeleton. After 1,000 days, the liver burden at the lower levels decreases with a half-time of about 8 years.

Placental transfer of ^{239}Pu in rats and mice has been reported to be inversely related to the size of dose given to the dam, (Finkel[241] in 1947, Wilkinson and Hoecker[242] in 1953) with maximum transfer of 3 to 8% of the dose, when the dose was given about 1 week after conception. Detectable amounts of ^{239}Pu were found in milk of mice, rats and cats after administering plutonium to the lactating dam. A more recent study by Ovcharenko[243] in 1971 showed that when pregnant rats were administered Pu-citrate intravenously on the day of birth and then allowed to give birth and nurse their young, the lactation did not significantly reduce the content of plutonium in the organs of the lactating mothers. Sacrifice of the mother rats after 1 and 10 days indicated 84.4% and 96.4% of the administered activity was retained in the body. The content of ^{239}Pu in the entire litter of baby rats fed by treated mothers from 1 to 10 days of age was 4.24 to 7.36% of the amount introduced. After 20 to 30 days 1.44 to 2.32% remained in the bodies of the baby rats.

In 1972 Sikov and Mahlum[244] reviewed the deposition and retention of plutonium in immature, developing animals. The absorption of ^{239}Pu from the gastrointestinal tract of juvenile rats and dogs relative to absorption in adults was shown graphically. As would be expected, the graph shows enhanced uptake of plutonium from the gut of the neonatal rat and dog is about 100-fold greater than in the adult of these species, with a gradual decrease to adult values to about the time of weaning.

Ballou[245] reported in 1958 that age appears to have a direct effect on absorption in rats. He noted 0.25% absorption of ^{239}Pu nitrate at 1 day of age, decreasing to 0.10% at 7 to 13 days of age, 0.02% at weaning and 0.003% in adult animals.

Finkel[246] in 1947 indicated that the absorption of plutonium bound to protein, as in milk, may be as much as 20-fold greater than in the uncombined form. This might increase the hazard to juveniles whose diet is primarily milk. On the other hand, McClellan[247] et al. in 1962 reported there are discrimination processes, which result in the concentration of plutonium in breast milk being substantially lower than that in the diet or the plamsa of the mother.

Sikov and Mahlum[244] indicated that there is a possibility that there is greater hazard to the gastrointestinal tract from ingestion of insoluble plutonium in the infant than in the adult. It has been shown that the gastrointestinal passage time of particles in juvenile rats does not differ from that in adults, although the average rate of passage is substantially reduced and there is evidence of prolonged retention in the stomach and ileum. This results in an increase in the average time that the plutonium is in contact with any given segment of the tract, and a corresponding increase in the radiation dose in the young, which is further increased because of the smaller organ diameters.

Sikov and Mahlum[244] also found that at 1 day after injection into adult rats, a much greater fraction of polymeric than of monomeric plutonium was localized in the liver. In the femur, the initial deposition of monomeric plutonium was much higher than that of polymeric. In the newborn there was an equal fraction of the administered dose of monomer and polymer in the femur at 1 day after injection and the concentration of the two forms in the liver were more similar than in the adult. With the passage of time, the monomer was removed from the liver of both age groups and the polymer was retained. This retention was more marked in the adults than in newborns, while the initial redistribution of plutonium from liver to bone was more marked in the juvenile rat than in the adult. As a result, at 30 days and again at 90 days, the patterns of partition of the two forms of plutonium were not markedly dissimilar in either the animals injected at birth or as adults. Buldakov[248] et al. in 1970 found similar patterns in studies on lambs.

The interpretation of data on immature animals is complicated by the fact that the size of individual organs or organ systems relative to that of the total body changes during development. Sikov and Mahlum[244] summarized the changes in the size of various organs with age. They also found that although the adult and newborn rat had a similar fraction of administered polymer in the liver at 1-day after ingestion, the corresponding concentration in the liver of the newborn was 20-fold that in the adult. In the adult, the concentration of polymeric plutonium in the liver decreased by about half in the first 90 days after injection; in the newborn, the dilution produced by growth resulted in an 80-fold decrease in concentration. At both ages there was a net increase in the fraction of the dose in the femur. This resulted in a slight increase in concentration in the adult, while there was a 20-fold decrease in the newborn. It is therefore evident that age at the time of injection influences the total radiation dose as well as the temporal distribution of the dose.

Incorporation of ^{239}Pu into the hair of rabbits was reported by Jaworowski[249] et al. in 1971. They found that when doses of 0.1, 1.0 and 10.0 µCi of ^{239}Pu chloride, dissolved in 0.5% phenol, pH 5.5, were injected subcutaneously into red-haired New Zealand rabbits, the mean incorporation of ^{239}Pu in the anagenetic hair of the first fleece (grown 22 days after dosing) ranged from 1.22 X 10^{-3} to 2.73 X 10^{-3}% of the dose per gram of hair. There appeared to be an inverse relationship of percent incorporation to dose amounts. Twice as much was incorporated at the 0.1 µCi per animal, as at 1.0 and 10.0 μCi per animal. In the growth of the second fleece, the rate of incorporation was 10 times lower at 10.0 μCi than at the lower doses. At the dose levels studied, the amount in hair was similar to that in muscle but much lower than bone or liver.

A recent report of the biological effects of plutonium in experimental animals was presented by Bair[250] in 1975.

BIOLOGICAL BEHAVIOR OF INHALED PLUTONIUM IN ANIMALS

Airborne plutonium particles are similar to most other particles when they are inhaled in that deposition in the respiratory tract is primarily dependent upon the physical properties of the particles and the respiratory characteristics of the individual inhaling the particles. After deposition in the respiratory tract, plutonium may be retained in the lung for a long time, be translocated to other tissues in the body, accumulated in the lymph nodes associated with the respiratory tract or excreted in urine and feces. The actual deposition of inhaled plutonium depends largely upon the physical and chemical characteristics of the inhaled material (Bair[251] and Lafuma[252]).

Bair[251] reviewed the literature in 1974 and presented figures and tables showing lung clearance and translocation values of several investigators. He notes that within the first week after exposure, a fraction of the deposited plutonium is cleared from the respiratory tract and excreted. In the case of PuO_2 inhaled by beagle dogs, all plutonium cleared from the respiratory tract is excreted in feces except for a small fraction that may be solubilized, absorbed into the circulating blood, and either cleared by the kidneys or deposited (by the blood) in another tissue. The magnitude of the fraction cleared within the first week depends upon the fraction of readily soluble material present and also upon the distribution of the deposited plutonium within the respiratory tract. Plutonium particles deposited on the ciliated epithelium of the upper respiratory tract are trapped in mucus, propelled to the esophagus and swallowed. Plutonium deposited in the lower regions of the lung, in the alveoli, is not available for clearance and may be incorporated into the cellular structure of the lung and retained for a long time. The clearance of plutonium deposited in the lower lung is generally assumed to be exponential over a reasonably long period of time.

Bair[251] gives retention half-times of several plutonium compounds. The retention half-times for organic complexes of plutonium, plutonium nitrate and fluoride range from less than 100 days to about 300 days in rats and dogs. The retention half-times for PuO_2 are substanially longer, ranging from 200 to 500 days in rats, and from 300 to 1000 days in dogs. The wide range of values observed in dogs is largely due to extensive experimentation with a variety of plutonium oxides with different particle size characteristics. Studies with $^{238}PuO_2$ in dogs indicate a much shorter lung retention time than is observed for $^{239}PuO_2$. This appears to be due to instability of $^{238}PuO_2$ particles, possibly caused by radiolysis in tissue fluids.

In 1970 Bair[253] presented figures showing the relationship of particle size and isotope on retention of plutonium in lung as well as for oxides prepared under different conditions. Oxide prepared by calcining the oxalate at 1000° C was retained with a half-time of 650 to 950 days compared with 300 to 400 days for the oxalate calcined at 350° C. Oxides prepared from metal powder at temperatures of 123° C to 450° C were retained in lung longer than the low-fired oxalate. Small particle size high-fired oxide was retained with a half-time of 400 to 500 days, compared with half-times up to 900 days for the larger particle size high-fired oxide. Retention of the small particle size $^{238}PuO_2$ was less than for the comparable $^{239}PuO_2$.

Plutonium may be removed from lung via bronchial and tracheal mucociliary processes and excreted but this accounts for only part of the plutonium cleared from the respiratory tract. Another fraction is accumulated by the lymphatics and deposited in lymph nodes. Data from an 11-year study with beagle dogs were presented by Park[254] et al. in 1972. After 5 years both lung and thoracic lymph nodes each contained 30% of the plutonium initially deposited in the lower respiratory tract. After 11 years the lung burden had decreased to about 9% and the thoracic lymph nodes had increased to 40%. Translocation of plutonium from lung resulted in levels in liver of about 10% of the alveolar-deposited plutonium, in bone of about 5% and in the abdominal lymph nodes of about 7%.

The relative distribution among body tissues of plutonium translocated from lung is essentially the same for all plutonium compounds. In beagle dogs, within several months after inhalation of plutonium nitrate, the fraction remaining in lung decreased to 40% or less of the amount deposited in the lower respiratory tract according to Ballou[230] et al. in 1972.

Translocation from the lung resulted in bone accumulating about 30% and liver about 10% of the alveolar-deposited plutonium. A small percentage was found in spleen, lymph nodes, and the soft tissues and the remainder was excreted in urine and feces.

Data presented by Park[255] et al. in 1973 showed that $^{238}PuO_2$ may be cleared from lung more rapidly than $^{239}PuO_2$. It has also been found that translocation of ^{239}Pu from lung to other tissues in the body may be greater than for ^{238}Pu.

Distributions of plutonium in tissues of beagle dogs 5 years after inhalation of $^{238}PuO_2$ and $^{239}PuO_2$ are also compared by Park[255] et al. Both oxides were calcined at 350° C. The particle size of $^{238}PuO_2$ was 0.1 μm (CMD) and of $^{239}PuO_2$ was 0.1 to 0.5 μm (CMD). The ultrafilterability was 1 to 2% for the $^{238}PuO_2$ and <1% for the $^{239}PuO_2$. After 5 years, only 10% of the body burden of the ^{238}Pu was in lung compared with 46% for ^{239}Pu. Accumulation in thoracic lymph nodes was 3 times greater for ^{239}Pu than for ^{238}Pu, however, the bone burden of ^{238}Pu was 12 times that of ^{239}Pu. This illustrates that the behavior of $^{238}PuO_2$ in the body may differ significantly from that of $^{239}PuO_2$.

In 1972 Craig[256] et al. exposed beagle dogs to $^{239}PuO_2$ aerosols as part of a low-level effects study of the oxides of both ^{238}Pu and ^{239}Pu. The alveolar deposition of $^{239}PuO_2$ aerosols was measured as a function of the aerodynamic particle size distribution of the aerosol and the tidal volume of the dog during exposure.

Matsuoka[257] et al. studied the distribution of plutonium nitrate aerosol by rats. Rats sacrificed immediately after exposure showed deposition in the tracheo-bronchial and pulmonary region. One day after exposure, plutonium was mainly in the lung with very small amounts in the intestine. One month after inhalation, the plutonium deposited in the lungs still remained, with very small amounts translocated to the trabecular bone of the vertebra and to the ileum. Isolated lung specimens showed no deposition in the tracheo-bronchial region and very little deposition in the pulmonary region.

In 1972 Suzuki[258] et al. also exposed rats to submicron aerosols of plutonium nitrate and determined the retention and excretion. Excretion of plutonium was mainly observed in the feces on the first day following inhalation and the daily fecal excretion decreased to 1/40 to 1/100 of the total intake of plutonium on the third day and thereafter. The urinary excretion was less than 1/10 of that of the feces.

Ballou[259] et al. studied the deposition of inhaled ^{239}Pu citrate in dogs and found that ^{239}Pu reached a peak concentration in blood after 6 hours, then decreased with clearance kinetics similar to intravenous administration. One day after exposure the lung, skeleton, liver, blood and intestines retained most of the inhaled ^{239}Pu. After 100 days 36% of the initial deposit was retained; 33% in the lung, 44% in the skeleton and 17% in the liver. Although the concentration in specific lymph nodes (hepatic, splenic, and tracheo-bronchial) was among the highest of any tissues analyzed, the total amount deposited in a selection of 12 lymph nodes (~10 g tissue) was always less than 0.6% of the body burden.

Wandall[260] reviewed the mechanisms involved in the lung clearance of inhaled $^{238}PuO_2$ and $^{239}PuO_2$ aerosols in dogs and rats and the subsequent translocation of the solubilized fraction throughout the body.

Dionne and Sanders[261] found that $^{239}PuO_2$ particles inhaled by rats were deposited in alveoli and rapidly phagocytized and concentrated within pulmonary macrophages. A hybrid computer alveolar model was developed in order to demonstrate the distribution of alpha energy in the alveoli and the modifying influence of pulmonary macrophages on the alveolar alpha-energy from phagocytized particles.

BONE DEPOSITION

Atherton[262] et al. administered ^{239}Pu(IV) intravenously to beagles in: (1) the Pu-transferrin complex, (2) the 0.08 M citrate buffer and (3) as a suspension of near colloid size particles at pH ~6. The skeletal retention of plutonium, when injected in a monomeric form, is 50% of the injected dose. The retention of polymeric plutonium in the skeleton was less than 1/20 that seen in monomeric or transferrin complexed plutonium at comparable times. The uniformity of skeletal distribution of polymeric plutonium from bone to bone is markedly less than that seen with the two monomeric forms. Reduced skeletal retention in animals injected with polymeric plutonium indicates a different skeletal retention mechanism in animals injected with polymeric plutonium. The authors postulated that skeletal retention of the polymer is not associated with bone per se but likely increases with the amount of red marrow a bone contains.

Stover[263] et al. reported the retentions of plutonium in the humerus and third lumbar vertebra of dogs injected with 0.016, 0.048, and 0.095 μCi/kg ^{239}Pu. The dogs survived from 1600 to 5200 days after injection. The data show that the rate of decrease of retention is greater during the first 2 years than at later times. There appear to be two phases in the time dependence of skeletal retention. Two things are happening during the first years after injection. Both the rates of the remodeling processes which remove plutonium from the bone surfaces and the fraction of skeletal plutonium that remains on the surfaces are decreasing. Both of these factors contribute to a decreasing loss of plutonium from the skeleton. The remaining plutonium which has been buried during the remodeling of the bone is much less accessible and is lost from the skeleton only at a very slow rate, if at all.

Another 1972 report by Stover[264] et al. compared the skeletal and hepatic dose rates from intravenously injected dogs. Young adult dogs were administered 0.0059 μCi ^{239}Pu(IV)/kg in 0.08 M citrate buffer, pH 3.5. The dogs were sacrificed at 1648 and 2546 days. The cumulative rad doses to the skeleton and liver were compared and the results show that the average dose to the liver exceeds that to the skeleton, except at the 2.8 μCi/kg dose level, after 1300 days.

In 1972 Jee[149] found that the effectiveness of the various routes of administration in delivering plutonium to bone, in decreasing order is: intravenous≳intraperitoneal>subcutaneous>intramuscular>inhalation>oral>direct application upon skin. Reticuloendothelial cells in the marrow compete with bone for polymeric plutonium and thus decreases the plutonium available for bone surfaces. The uptake in young and adult bones differed by a factor of two. The fate of the plutonium surface deposits is modified by bone growth, modeling and remodeling in growing animals and remodeling in adults. These age-related processes remove the plutonium from bone surfaces and/or bury the surface deposits with new bone.

A very complete summary of the metabolism of plutonium after administration in different forms and by different routes up to 1972, is given in tabular form in the International Commission on Radiological Protection (ICRP) Publication 19[265].

BIOLOGICAL EFFECTS OF PLUTONIUM IN ANIMALS

In animal experimentation, the acute forms of toxicity are manifested by an irradiation syndrome comparable to that produced by a total irradiation with x-rays. Forms of a chronic nature can produce broncho-alveolar tumors in the lung, bone cancers, and bone fractures in the skeleton, and, with less frequency, liver tumors. Saenz and Ramos[1] present lethal and tumor-causing doses of plutonium in dogs, rabbits and mice.

It has been shown that the distribution of plutonium and the radiation dose among the tissues in the body will vary depending upon the physical and chemical characteristics of the plutonium inhaled. The biological effects that occur will depend upon the radiation exposure and the relative radiation sensitivity of each tissue into which plutonium is transported and deposited. These are primarily blood, bone, liver, lung and lymphatic system.

Blood

Plutonium is cleared from the circulating blood within a few hours after absorption. Therefore, the effects seen in blood cells are probably due to irradiation of hematopoietic tissue in which plutonium is deposited or to irradiation of blood circulating through plutonium-containing tissues.

In 1972 Dougherty[266] reported blood cell alterations in dogs receiving a single intravenous injection of a wide range of dose of ^{239}Pu (0.016 to 2.8 μCi/kg). Dogs receiving the four highest dose levels showed changes that were dose-dependent and were most marked in granular leukocytes which were maximally depressed at 2 to 3 weeks post injection. A sustained lymphopenia occurred at the two highest dose levels. A transient early thrombocytopenia and anemia were also found in higher level dogs. A moderate anemia reappeared as the animals became terminal.

In 1972 Nabors[267] noted that beagles bearing ^{239}Pu burdens show the most marked alterations in serum alkaline phosphatase and serum glutamic oxaloacetic transaminase and serum glutamic pyruvic transaminase. Animals with low dose levels appear to have a much longer latent period prior to the appearance of altered serum biochemistry than do animals with higher dose levels.

Bone

The most sensitive effect of plutonium deposited in bone is radiation-induced osteogenic sarcoma. Because plutonium deposits on bone surfaces, a large fraction of the alpha energy is absorbed in sensitive cells. Although osteogenic sarcoma has been reported in rats that accumulate doses of less than 10 rads, statistically significant increases in tumor incidence have not occurred at doses less than 30 to 50 rads (Bair[250]). The available data suggest that the dose-effect curve for dogs is different from that for rodents. Mays and Lloyd[268] conclude that the incidence of plutonium-induced osteogenic sarcoma is 0.38%/rad for beagle dogs, 0.10%/rad for mice and 0.06%/rad for rats.

No bone sarcomas have been reported in dogs following $^{239}PuO_2$ inhalation but they are the important late biological effects in dogs which have survived intravenous injection of plutonium citrate for more than 3 or 4 years (Mays[269] et al.).

Osteogenic sarcoma has been observed in mice, rats, rabbits, and dogs after intravenous injection of several ^{239}Pu compounds including organic complexes and plutonium nitrates.

Osteogenic sarcoma has also been observed in rats after inhalation of $^{238}PuO_2$, but has not been reported in any animal species after inhalation of $^{239}PuO_2$.

In 1973 Moskalev and Strelt'sova[270] found that optimun osteosarcomogenic doses to rat skeleton were in the range of 0.7 to 1.8 krads for ^{239}Pu. The osteosarcoma incidence was identical for both male and female rats but significantly age dependent for each sex.

Pesternikov and Bukhtoyarova[271] discussed the quantitative assessment of osteosarcoma growth rate in rats following a single intraperitoneal administration of 2.5 mCi/kg of ^{239}Pu citrate. Osteosarcomas are shown not to predominantly originate at the sites of the highest isotope concentration. Of some tumors, primary multiple growth was characteristic. Possible extrapolation for determining the osteosarcomas development time was noted.

James[272] reported a study in which 6 to 8 week-old rats were given a single injection of 4.5 μCi/kg soluble $Pu(NO_3)_4$, a dose level expected to give an approximate 80% yield of osteosarcomas. Localized dose rates were measured at fixed reference distances between 5 to 20 μm from bone surfaces by counting gamma-flux entering small cylindrical targets located in a thin nuclear emulsion in contact with a bone section. Dose rates 1 day after the injection ranged from 22 to 57, 12 to 37, and 7 to 26 rads/day at distances of 5, 12.5, and 20 μm, respectively, from a number of different endosteal surfaces. These local dose rates changed with time by factors of the order of 2, in either direction, depending on the prevailing conditions of remodeling. A cumulative alpha dose of about 2000 rads (delivered to primitive cells at trabecular surfaces over a period of 36 weeks) may be associated with a 10% probability of developing a tumor in an individual femoral epiphysis. Taylor[273] et al. studied the general syndrome induced by ^{239}Pu(IV) injected intravenously in beagle dogs. The most critical factors were the induction of bone cancer, hematopoietic changes and liver lesions. Some of the less serious end points were pathological fractures, dental changes and atrophy of the turbinates. These latter conditions produced functional impairment in only part of the dogs and principally at the higher dose levels.

Taylor and Mays[274] gave young adult St. Bernard dogs a single intravenous injection of ^{239}Pu, because of their high spontaneous incidence of bone cancer. The appearance time of radiation induced osteosarcomas in this giant breed was approximately one-half of that observed in beagles when both were injected with 0.3 μCi ^{239}Pu/kg of body weight.

In 1972 Moskalev[275] reviewed the results of studies by Soviet scientists on problems of the biological action of ^{239}Pu. Main attention was given to the analysis of late effects (tumor and nontumor effects) developing in the body as a result of injury by ^{239}Pu, dose-effect curves for bone and lung tumors, estimation of minimum carcinogenic dose levels, and determination of doses not affecting the natural life span.

Liver

Liver accumulates levels of plutonium similar to bone. However, liver tumors have not been a common finding in experimental animal studies. Bile duct tumors have been observed to occur in beagle dogs at doses as low as 60 rads. However, not only was the incidence very low, but bile duct tumors also occurred in control dogs (Taylor[276] et al.).

Kashima[277] et al. demonstrated in 1973 that liver function at 21 days post exposure was markedly reduced in rats injected with ^{239}Pu at birth or at 7 days of age. The differing effects produced by monomeric and polymeric plutonium were explained by differences in radiation dose to the liver.

Lung

Inhalation of relatively soluble plutonium compounds, such as organic complexes, plutonium nitrate and plutonium oxide, has resulted in primary pulmonary neoplasia in rodents, rabbits and dogs (Bair[251]).

Pulmonary neoplasia has also been observed in beagle dogs, baboons (Metivier[278] et al.) and rodents after inhalation of $^{239}PuO_2$. Tumors were observed in animals that had lung doses less than 10 rads. Statistically significant increases in pulmonary neoplasia occurred at doses of about 30 rads and above. In several experiments, increased incidences of lung tumors occurred at doses between 10 and 100 rads. In an 11-year study of $^{239}PuO_2$ in beagle dogs, nearly all animals which had depositions of between about 3 nCi and 50 nCi per gram of lung had lung tumors. Dogs which had a deposit of more than 50 nCi per gram of lung died early due to radiation pneumonitis and fibrosis (Park[254] et al.). The mean dose to the lungs of the dogs that developed neoplasia ranged from 1200 to 4000 rads. As in the case of osteogenic sarcoma, the dose-effect curve for pulmonary neoplasia in dogs appears to differ from the rodent dose-effect curve.

Howard[279] described the results of a long-term study to evaluate the pathologic effects in the lungs of beagle dogs following $^{239}PuO_2$ particle inhalation by 40 dogs exposed to $^{239}PuO_2$ particles having a CMD of 0.1 to 0.5 μm; 22 died with primary lung neoplasia 38 to 110 months post exposure; 8 died of pulmonary fibrosis; 5 were sacrificed for radionuclide distribution measurements; and 5 were still alive over 9 years post exposure. Initial alveolar deposition ranged from 0.5 to 3.5 μCi and accumulated radiation dose was 2500 to 12,000 rads. Most lung tumors were found to be bronchial-alveolar carcinomas of peripheral origin, with two peripheral squamous cell carcinomas and three epidermoid carcinomas. Also in this group of dogs there were three thoracic sarcomas and two with malignant lymphomas.

Sanders and Park[280] and Sanders[281,282] reported on the carcinogenicity of inhaled ^{238}Pu and ^{239}Pu in soluble and insoluble forms by rats and dogs. It was concluded that exposure of rats to even small amounts of ^{238}Pu resulted in significant incidence of tumors in lungs. The type of metaplasia and neoplasia in rats and dogs indicates the epithelial, either alveolar or bronchiolar, origin of most of the observed tumors.

Anderson[283] et al. reported on lung irradiation of Syrian (golden) hamsters with static plutonium microspheres. Exposure was accomplished by injecting 10 μm microspheres into the jugular vein. The microspheres were trapped in the capillary bed of the lung. Activity range was 0.01 to 59 pCi/sphere. Of 1,142 hamsters exposed to 5,700,000 microspheres, only two lung tumors were observed. Although the experiment was not complete at the time of the report the authors felt that the volume of tissue irradiated was an important factor and that locally concentrated alpha irradiation is less damaging in the hamster lung than the same amount of energy delivered over larger volumes.

Lymph

Plutonium accumulates in lymph nodes following deposition of plutonium in the respiratory tract. Months or years after the contaminating event, lymph nodes may attain concentrations of plutonium many times the average concentrations remaining at the site of deposition and consequently the accumulated radiation dose to lymph nodes may be greater than to any other tissue.

Although dogs have been studied for 11 years after inhaling a variety of plutonium compounds, primary neoplasia of lymphatic tissue has not been a common observation (Bair[253]). Metastases of tumors from lung to lymph nodes have occurred, but none has been the primary cause of death. It can be concluded from the many rodent and dog life-span experiments with ^{238}Pu and ^{239}Pu, in which many lymph nodes have been exposed to doses ranging from near background to thousands of rads delivered at a wide range of dose rates, that the lymph nodes are not especially susceptible to the carcinogenic action of alpha radiation from plutonium.

Dagle[284] reported on lymph node clearance of ^{239}PuO$_2$ administered as insoluble particles from subcutaneous implants in adult beagles to simulate accidental contamination of hand wounds. The popliteal lymph nodes were monitored following implantation ranging from 9.2 to 39.4 μCi ^{239}PuO$_2$ into the hind paw. Groups of dogs were sacrificed 4, 8, 16, and 32 weeks later and it was found that the popliteal lymph node contained from 1 to 10% of of the implant dose. Histopathologic changes in the popliteal lymph nodes with plutonium particles were characterized primarily by reticular cell hyperplasia, increased numbers of macrophages, necrosis and fibroplasia. Eventually, the plutonium particles became sequestered by scar tissue that often replaced the entire architecture of the lymph node. There was slight clearance of plutonium from the popliteal lymph nodes of dogs monitored for 32 weeks.

Other Effects

Stover[285] et al. found correlation between dose level and life-shortening in beagle dogs. Sikov and Mahlum[244] found that 12 μCi/kg of ^{239}Pu, which is well below the acute lethal range for the adult rat, produces extensive prenatal death within 3 to 5 days when administered to the pregnant rat at 9 days of gestation. There was evidence that these deaths occur by alterations of the nutritive function of the fetal membranes. In contrast, no prenatal mortality is produced at doses beyond the lethal level for the mother, 150 μCi/kg, if administered at 15 or 19 days of gestation. In 1972 Ovcharenko[286] found similar results when studying the effects of plutonium on reproductive ability of rats. He found that the character and depth of radiation-induced changes depended upon the amount of activity injected and the time elapsed since injection.

In the course of investigating the postulated development of the offspring of experimental animals, a number of peculiarities were observed: decrease in viability, delayed physical development, variations in weight, disturbance of blood formation, change in radiosensitivity and depression of sex function.

Sanders[287,288] and Sanders and Jackson[289] studied the synergistic effects of plutonium and asbestos administered to rats. It was found that coadministration by inhalation of ^{239}PuO$_2$ with asbestos resulted in less excretion of ^{239}Pu than was the case with ^{239}PuO$_2$ only. Focal granulomatous lesions were common around bronchiolar areas of the lung in animals receiving asbestos. Plutonium oxide particles tended to concentrate in these asbestos-induced lesions forming hot spots of alpha activity. When the same combination was intra-abdominally injected into rats, asbestos acted in an additive manner with $^{239}PuO_2$ in inducing mesotheliomas, the two agents combined having an effect equal to the sum of their effects when administered separately. A similar additive response was found when benzpyrene and $^{239}PuO_2$ were injected with resulting formation of abdominal sarcomas.

COUNTERMEASURES FOR PLUTONIUM IN ANIMALS

A number of chelating agents have been used in experiments to remove ^{239}Pu by increasing the excretion rate. Smith[290], as reported in 1958, testing 10 of the amino-acetic acid-type chelators administered intraperitoneally in a dose of 1.5 mM/kg in an equivalent amount of calcium gluconate 1 hour after intravenous injection of ^{239}Pu to rats. The most effective agent, diethylene-triamino-pentaacetic acid (DTPA), decreased deposition of plutonium in the liver and bone by about 99%; the next most effective, diamino-diethylether-tetraacetic acid (DDETA), reduced deposition by about 95%. Even when DTPA calcium gluconate plus 25,000 IU of Vitamin A was administered 39 days after a dose of ^{239}Pu, it reduced retention in bone by 25% and in liver by 80%.

Zirconium citrate and certain phosphate compounds, especially hexametaphosphate (HMP) reduced retained ^{239}Pu by 50 to 70% if administered immediately after a dose of ^{239}Pu according to Wagner and Temple[201] and Rosenthal[292]. However, all of the above mentioned treatments have certain drawbacks: DTPA and its analogs increased ^{239}Pu concentration in the kidney, while zirconium and phosphates increased concentration in the liver. Desferriox-amine (DFOA) appears to be as effective or nearly as effective as DTPA if given within a few hours after ^{239}Pu exposure; it did not show significant ^{239}Pu renal concentration according to Taylor[293].

Nenot[294] et al. found that the therapeutic effectiveness of DTPA on bone deposits of plutonium can be appreciable. DTPA treatment begun 3 weeks following intramuscular injection of ^{239}Pu decreased bone burdens by a factor of one-half to one-third in 3 months.

Pretreatment of rats with tetracycline was found to decrease retention of ^{239}Pu in bone to about 60% of control values according to Taylor[295] et al. Tetracycline increased the initial rate of clearance of ^{239}Pu from plasma. Treatment with tetracycline was found to have no effect on the distribution of intramuscularly-injected ^{239}Pu, even when treatment was started immediately after contamination.

Morin[231] et al. studied the effect of treatment by DTPA following intramuscular and inhaled ^{238}Pu nitrate by rats. Treatment was initiated 24 hours after exposure and repeated twice a week. Protection of or removal from bone by DTPA was clearly demonstrated at the 30th day when the respective bone burdens of control and treated animals were in a ratio of 8:1, a decrease from 45 to 17% of bone activity. Postponing treatment until the 21st day showed the importance of early treatment.

Sanders[296] and Sanders and Meier[297] studied the influence of fasting on the efficiency of DTPA in enhancing the excretion of intraperitoneally injected ^{238}Pu in female rats. Complete fasting for 10 days had no significant effect on the cumulative excretion of injected ^{238}Pu. In animals given DTPA within the first hour after ^{238}Pu there was a 2.2-fold increased excretion, and a 1.4-fold increased excretion when DTPA was administered 14 days after ^{238}Pu, irrespective of fasting. They concluded that distribution of ^{238}Pu bound to liquids in extracellular fluids and bone was not significantly altered by fasting so as to influence the chelation of ^{238}Pu or ^{239}Pu by DTPA.

Studies of the translocation of plutonium from wound sites and effects of treatments were reported by Bestline[298] et al., Lebel and Watters[299], Watters and Lebel[300], Watters and Johnson[301], and Gomez[302] et al. The investigators found that the effect of DTPA treatment was greater for $Pu(NO_3)_4$ than for PuO_2 implants. A time study of the buildup of plutonium in the lymph node draining the wound site disclosed that the lymphatic system provides rapid translocation from the wound. Rapid surgical excision, to be highly effective, should be performed as soon as possible. If accumulation of plutonium is detected in lymph nodes, lymphadenectomy should be considered.

Rosenthal[303] et al., Rosenthal[304] et al. and Baxter[305] et al. studied the effectiveness of DTPA and glucan, a polysaccharide derived from yeast cell walls, as agents for plutonium decorporation. DTPA therapy removed over 96% of intravenously injected monomeric ^{239}Pu from beagle dogs. Removal from spleen, lungs, kidneys, testes, muscle and lymph nodes ranged between 50 to 90%. The total skeletal content was also reduced by 50%. Adjunct therapy with glucan did not result in significant additional removal of plutonium. Use of the same treatment following polymeric ^{239}Pu injection of mice showed that at 47 days after administration, the net removal was 8.5% of the injected dose by glucan, above 11.5% by twice weekly DTPA treatments and 19.5% by combined therapy.

In 1972 Ballou and Hess[306] found that about 50% of the plutonium excreted into the perfused intestine of the rat during the first hour after plutonium injection arrived by way of the bile. After DTPA treatment, the proportion excreted in bile was increased to 75% and the bile plutonium concentration increased 15 to 20-fold.

Rahman[307] et al. in 1973, successfully encapsulated EDTA and DTPA within lipid spherules (liposomes). Encapsulated EDTA given intravenously to mice was retained longer in tissues than nonencapsulated EDTA. Encapsulated DTPA, given to mice 3 days after ^{239}Pu injection, removed an additional fraction of plutonium in the liver, presumably intracellular, not available to nonencapsulated DTPA. It also increased urinary excretion of plutonium.

Inhaled insoluble plutonium is not effectively mobilized by a wide variety of expectorants, mucolytes agents, bronchodilators, surface-active agents, phagocytic stimulating procedures, chelating and other physiologically active materials (Smith[308]). Recent experiments have shown that pulmonary lavage with physiological saline may remove 50% or more of PuO_2 from the lung of rats [(Sanders[309]); dog and rat (McDonald[310]); monkeys (Kunzle-Lutz[311] et al., Nolibe[312]); and baboons (Kunzle-Lutz[313] et al.)].

DEPOSITION OF PLUTONIUM IN MAN

Plutonium has found its way to man, in readily measurable quantities, only through occupational exposure, where the route is usually direct--by ingestion, inhalation, or by way of a plutonium-contaminated wound (Bair and Thompson[314]).

In 1959 Langham[315] found that alpha radiation from plutonium on the skin surface does not penetrate to the sensitive basal layer of the epithelium. Absorption of plutonium through the skin occurs only to a very slight degree, and probably only when the skin is damaged. Lister[316] studied fecal and urinary plutonium excretion from two subjects who sustained high levels of contamination on the uncut skin of the hand from accidental contact with acid plutonium solutions. In one case, where the contaminant was mixed plutonium isotopes in aqua regia and nitric acid, the measurements covered 150 days. In the other case, contamination with a solution of plutonium in dilute hydrochloric acid containing EDTA and a detergent was followed for 110 days. The excretion patterns showed marked differences from those found by Langham[315], particularly in the high and variable amount of plutonium excreted in feces relative to urine.

Laylee[317] also studied excretion of plutonium following a lacerated wound contaminated with 100 nμCi of PuO_2. No translocation values were given.

In October 1970, a 59-year-old employee of Dow Chemical Company died from complications following open heart surgery. This employee had an estimated systemic burden of 0.22 μCi of plutonium or 541% of the maximum permissible system burden, at the time of his death. The employee had been involved in several contamination incidents and had two contaminated puncture wounds prior to November 1965. He had excreted measurable amounts of plutonium in his urine since 1960 and had an estimated 21% of the maximum permissible systemic burden. In July 1965, he received another puncture wound which left 18.5 μg of plutonium. A surgical incision removed 5.9 μg in 1967, a second incision removed all but about 0.3 μg. Autopsy and tissue analysis showed plutonium concentration>skeleton>liver>lung>tracheobronchial lymph nodes>spleen>kidneys. The total extrapolated system burden from autopsy samples was 0.07 μCi or about 177% of the maximum permissible amount (Lagerquist[318] et al.).

In 1973, Popplewell[319] reviewed the chemistry of plutonium incorporated in the body. Subjects covered are transport within the body in relation to use of chelates, especially Ca-DTPA; studies of transferrin complexes in body fluids; intracellular uptake in liver and deposits in bone.

Keely and Wenstrand[320] calculated the urine to fecal ratios for two workers following inhalation of soluble plutonium nitrate. Twenty days post exposure the residual body burden was primarily in the skeleton. After 80 days and 250 days post-exposure <10 nCi was in the skeleton and <2 nCi total plutonium in liver or lung as determined by in vivo counts. Urinary excretion was erratic after 20 days post exposure but indicated a 1.8-day half-life during the initial time of translocation.

Lagerquist[321] et al. studied 19 cases where tissues were obtained from autopsy and analyzed for plutonium. Twelve of the cases had no known record of plutonium exposure. All of these had some detectable plutonium in their organs but generally an amount that is representative of less than 1% of the Maximum Permissible Systemic Burden (MPSB). In the seven others, the distribution varied from case to case, depending on the mode of entry, the chemical form and the length of time since exposure. A typical distribution was not found, but on the average, the lung and tracheo-bronchial lymph nodes had the highest concentration of material followed by the liver, the bones, and other tissues.

Przyborowski[322] and Vaane[323] et al. studied the deposition of plutonium particles in the human respiratory tract after inhalation. A combined experimental mathematical procedure was used.

McInroy[324] et al. studied autopsy samples from 70 former employees of Los Alamos Scientific Laboratory who had exposures ranging from 4 to 30 years. Exposure in most cases was to inhaled PuO_2 aerosols. Thirty-three of the measured cases had plutonium deposition in th tracheobronchial lymph nodes. No abnormalities of the lymph nodes were found.

Richmond[325] presented a summary of information on human experience with plutonium. Groups studied ranged from exposed GI's in 1944 to 1945 weapons development, to occupationally exposed workers at Los Alamos and Rocky Flats, to inhalation intake and burden in man of fallout ^{239}Pu in New York. Data are presented in tabular and graphical forms with tissue distribution included.

The retention half-time for PuO_2 inhalation in humans was reported as 250 to 300 days. The relatively low values for human beings, compared with dogs, suggest either that man clears plutonium particles from his lungs more rapidly than dogs or that the materials inhaled in human accident cases were more soluble than plutonium dioxide (Bair[326]).

Durbin[327] reviewed old data and applied more recent knowledge derived from animal experiments. Much of the older information was derived from intravenous administration of tracer amounts of ^{239}Pu citrate to men suffering incurable diseases in 1945 and 1946. A few days after injection, human soft tissues (other than blood and liver) contained as much as 20% of the plutonium dose. Five to 15 months after injection the average liver plutonium content was 31% of the dose for three cases with presumably normal liver function. Four to 457 days after injection, mean total skeletal plutonium was 49% for the seven cases judged to have most nearly normal liver and skeletons.

Plutonium is transported in blood combined with transferrin, the iron transport protein, and is stored in the liver in association with stored iron. After being bound to transferrin, plutonium partially traces the behavior of the carrier protein. The early phases of plutonium transport which are apparently associated with extracellular fluid mixing, were prolonged in individuals with impaired circulation.

Maximum urinary plutonium excretion occurred before the bulk of plutonium was protein-bound. Minimum urinary excretion coincided with the time of maximum Pu-transferrin binding. These observations were taken to mean that some plutonium is filtered by the kidney in the form of a low-molecular weight chelate. Urinary plutonium excretion was reduced by one-half in those persons who were anemic, presumably because of their more efficient Pu-transferrin binding.

Fecal excretion of plutonium apparently represents secretion in bile and other digestive juices. Fecal excretion was reduced by one-half or more in those persons whose gastrointestinal tracts were judged not to be normally stimulated.

Average soft tissue release half-time was estimated to be not less than 480 days and bone surface turnover for the whole adult human skeleton was estimated to be about 5% per year. For an individual on a diet adequate in iron and with normal iron stores, the model predicts that bone and liver will contain equal amounts of plutonium 15 years after exposure.

EFFECTS OF PLUTONIUM IN HUMANS

Hemplemann[328] et al., reporting a 27-year study of Los Alamos workers, found that to date, none of the medical findings in the group can be attributed definitely to internally deposited plutonium. The bronchial cells of several of the subjects showed moderate to marked metaplastic change, but the significance of these changes is not clear. Diseases and physical changes characteristic of a male population entering its sixth decade were observed. Because of the small body burdens, on the order of the maximum permissible level, in these men so heavily exposed to plutonium compounds, it was concluded that the body has protective mechanisms that are effective in discriminating against these materials following some types of occupational exposures. This is presumably explained by the insolubility of many of its compounds. Plutonium is more toxic than radium if deposited in certain body tissues, especially bone; however, from the practical point of view, plutonium seems to be less hazardous to handle.

In 1974 Tamplin and Cochran[329] reported two cases of cancer presumably caused by plutonium contaminated puncture wounds of the hand. At this time these were the only reported cases of cancers in people working with plutonium.

COUNTERMEASURES FOR HUMANS EXPOSED TO PLUTONIUM CONTAMINATION

The most successful treatment of inhaled plutonium particles appears to be pulmonary lavage, followed by chelation therapy (Nolibe[330], Smith[331], McClellan[332] et al.). Lavage appears to remove about half the inhaled particulates. Chelation by means of administration of DTPA either by aerosol or intramuscular injection has been successful in removing soluble translocated plutonium (Nenot[333] et al. and Schofield and Lynn[334]).

Treatments of contaminated wounds include immediate use of a venous tourniquet; flushing and decontaminating the wound site; excising tissue; and administering chelating agents (intravenously, orally, and by aerosol inhalation) (Jolly[335] et al., Hesp and Ledgerwood[336] and Larson[337] et al.).

PRESENT ANALYTICAL METHODS

A thorough evaluation of present analytical methods for the transuranic elements including plutonium in environmental samples has been prepared by Bernhardt[338] and is in press at this time.

It has been noted by numerous authors that analytical results are only as good as the methods by which the samples are collected. Among the environmental samples posing the greatest sampling problems are air and soil. In 1972 Hull[339] reported tentative methods for the sampling and analysis of radioactive substances in air as formulated by the Radioactive Substances

Subcommittee of the Intersociety Committee on Methods for Air Sampling and Analysis.

In 1973 Eberhardt and Gilbert[340] discussed the problems associated with sampling of soils for environmental levels of plutonium. The general statistical considerations of sampling, problems associated with compositing samples and consistent use of random sampling as a basic technique were examined.

The literature contains many methods for determination of plutonium in biological and environmental samples. Most of these methods are variations of basic procedures which utilize the precipitation, ion exchange and complex forming properties of plutonium followed by electroplating and counting.

GENERAL METHODS

In 1971 Wong[341] reported on the radiochemical determination of plutonium in seawater sediments, and marine organisms. Seawater samples of 50 to 60 liters were collected at various depths; 50 to 100 g of dried shallow water sediments or a larger aliquot from core samples was used. The author provides a table of average ^{239}Pu concentrations in marine organisms to allow selection of proper sample size for optimal sample counting rate; the activity should be about 0.5 to 5 dpm. An iron(II) hydroxide coprecipitation method was used for the concentration of plutonium in seawater. A nitric-hydrochloric acid leaching method was adapted for the treatment of sediments and ashed organisms. The sensitivity for this method is 0.004 dpm per 100 liters of seawater (using 50-liter sample), 0.02 dpm per kg of sediments (100-g sample) and 0.002 dpm per kg of marine organisms (1-kg sample). Factors influencing the uncovering, contamination and blank activity were discussed.

Darrall[342] et al. described a method for the simultaneous determination of ^{241}Pu and plutonium alpha activity in effluent samples from nuclear installations. The plutonium was isolated by coprecipitation on barium sulfate followed by extraction into di-(2-ethylhexyl)-phosphoric acid, which was incorporated in a liquid scintillator for counting in a liquid scintillation spectrometer. Interferences from alpha- and beta-emitting radionuclides were studied together with interferences from nonradioactive elements. The lower limit of detection is in the region of 1 pCi. This is a modification of the method described by Sill[343] and by Sill and Williams[344] in 1969 who reported that plutonium will coprecipitate on barium sulfate provided it was in the tervalent or quadrivalent state and the coprecipitation was carried out in the presence of potassium ions. By coprecipitation after selective oxidation the elements can be separated. The barium sulfate precipitate may be alpha counted directly or the plutonium can be electrodeposited for alpha spectrometry. Talvitie[345,346] described methods for radiochemical determination of plutonium in environmental and biological samples by ion exchange and an electrodeposition method for alpha spectrometric determination. Sample preparation procedures were given for urine, animal tissue, bone, saline and nonsaline water, siliceous and limestone soil, and glass fiber air filters. Samples were prepared as azeotropic 6M hydrochloric acid solutions of Pu(IV), stabilized with hydrogen peroxide, and absorbed on anionic resin from 9M hydrochloric acid solution. Coadsorbed iron was removed from the resin with 7.2M nitric acid. Plutonium was selectively eluted with 1.2M hydrochloric acid - 0.6% hydrogen peroxide and electrodeposited from 1M ammonium sulfate at pH 2 for alpha spectrometric determination. Minimum detectable activity for 1000-min counts was 0.02 pCi

^{239}Pu. Puphal and Olson[347] describe a method for electrodepositing various alpha-emitting nuclides singly or in combination onto a stainless steel cathode from a mixed ammonium oxalate-ammonium chloride electrolyte. The nuclides were deposited as hydroxides to better than 98% in 50 minutes. Interferences are also discussed.

Gillette[348] et al. investigated the determination of plutonium in soil. The method involves a complete dissolution of the soil by a fusion technique followed by chemical separation of the plutonium by barium sulfate, extraction, electrodeposition, and alpha pulse height analysis.

Hampson and Tennant[349] reported the simultaneous determination of actinide nuclides in environmental materials contaminated by controlled discharges of liquid wastes. Multielement actinide analysis is achieved by extracting the whole group, or part of it, in the tri-n-octylphosphine oxide-n-heptone-nitric acid-sodium nitrate system, stripping into ammonium carbonate solution and electrodeposition, followed by solid-state alpha spectrometry. Up to 2 kg of edible seaweed or 10 kg of fish flesh can be handled, with detection limits (in terms of activity to double background) of 2×10^{-6} and 4×10^{-7} pCi/g, respectively, for a 1-week counting time. Sensitivities for precision with 4 percent standard deviation are 4×10^{-4} and 8×10^{-5} pCi/g, respectively, which corresponds to levels associated with fallout.

Piltingsrud and Stencel[350] report a direct method for evaluating ^{239}Pu content in large soil samples (up to 1 kg) by means of a phoswich detector. Sample ^{239}Pu + ^{241}Am specific activity must be >20 pCi/g. When high levels of other gamma emitters, a ratio >1000:1, are present causing analysis interference, specific radiochemical analysis of the sample is indicated.

In 1973 Thomas[351] presented a method for measuring ^{238}Pu and ^{239}Pu in air samples. The membrane filter was prepared by fusion and the plutonium was retained on an ion exchange column and eluted with HCl containing HI. After electroplating, the plutonium isotopes were alpha counted.

Novoselova[352] et al. described the optimum conditions of concentration, separation and measurement of ^{239}Pu after the latter had been added to ashed bone tissue. Bone tissue was ignited in a muffle furnace at 600° C, and the inorganic residue was dissolved in HNO_3. The plutonium was extracted with TBP, reextracted with aqueous hydroxylamine HCl, and precipitated from solution into bismuth phosphate. The latter was washed, mixed with a phosphor, and the plutonium was determined in a scintillation counter.

Ershova[353] et al. described a method for determination of ^{239}Pu based on liquid-liquid extraction by monoisoactyl methyl phosphonate and mixed isoamyl phosphate at a phase volume ratio of at least 100. This method is used for the estimation of the ^{239}Pu content in lung tissue, bone tissue, surface waters, and other environmental objects.

Ghysels[354] reported the extraction of plutonium was effected with liquid ion exchangers (e.g., tertiary amines such as triisooctylamine). The plutonium was put back into organic solution by extraction with DEHPA. After mixing with a liquid scintillator, the counting yield was determined (close to 100%). This method was intended for determination of plutonium in biological substances. It was applied to the analysis of radioactive dusts of different origin.

Schieferdecker[355] presented a method for separation of plutonium in pCi amounts from biological material by extraction with bis (2-ethyl-hexyl) phosphate (DEHPA). It was possible to identify individual transuranic elements after joint carrier-free preparation by electrodeposition and subsequent alpha spectrometry. Amounts below 0.2 pCi can be determined.

In 1973 Budnitz[356] provided an overview of the techniques for measuring plutonium in various media. The emphasis was on measurements for surveillance and protection in environmental and occupational situations. Overviews were first provided for the characteristics of, sources of, and typical levels of plutonium concentrations. The various measurement techniques were discussed.

AUTORADIOGRAPHY

Fission track autoradiography has been used by several investigators to study the distribution of plutonium in bone and other biological samples (Hamilton[357], Becker[358] et al., Jee[359,360] et al. and Schlenker and Oltman[361]). The autoradiographs are produced by placing thin bone sections in contact with plastic films which register fission fragment tracks. The bone and film are inserted into a nuclear reactor where neutrons induce fissioning of plutonium.

Some plastics show not only the fission tracks but an image of the bone itself. The image shows no detail but does reveal where the boundaries between the bone and marrow spaces lie so that positioning of the fission tracks relative to the boundaries may be easily determined. Lexan is the plastic which has been most widely used to produce images of irradiated materials (Schlenker and Oltman[361]).

Lindenbaum and Russell[362] found good agreement between liver burdens determined autoradiographically or radiochemically. Using livers from mice injected intravenously wiht polymeric ^{239}Pu, at dose levels ranging from 6.6 to 93 μCi/kg, the quantitative technique for assay of plutonium deposited in animal tissue was tested for linearity in the relationship between photographic exposure time and number of alpha tracks formed.

INSTRUMENTAL ANALYSIS

In 1973 Winkle and Hoetzi[363] reported a sensitivity of 0.04 to 0.01 pCi/m^3 for long-lived alpha-emitters in air. The sensitivity was obtained by alpha spectrometry of electrostatically-precipitated aerosols in a large Frisch grid ionization chamber after decay of the natural activity for 2 to 4 hours and 100 minutes counting. This sensitivity corresponds to about 0.1 to 0.01 of the maximum permissible concentrations (168/h/wk) for ^{239}Pu.

Liquid scintillation counting of plutonium in biological samples is the subject of a report by Joshima and Matsuoka[364]. The liquid scintillation method without ashing was established for plutonium in animal tissues by use of a commercially available tissue solubilizer. Samples up to 250 mg wet weight could be handled with this procedure. The samples were then counted in a liquid scintillation spectrometer set for the optimal gain setting for plutonium.

Matsuoka and Joshima[365] developed a method for determining particle size of plutonium in solution by a diffusion chamber. The diffusion chamber was prepared from a plastic ringe (20 mm ϕ x 5 mm), both sides of which were covered with millipore membrane of adequate pore size of Visking membrane. The diffusion chamber containing 0.1 ml of test solution was placed in a plastic container with 5 ml to 10 ml saline solution and incubated 1 hour with shaking in a 37° C water bath. The "diffusibility" of plutonium particles from the diffusion chamber was estimated from the sample measurement of extra fluid at the final point by liquid scintillation counting and for external counting at 17 keV L-x-ray of the diffusion chamber before and after the incubation. The particle size of "monomeric" and polymeric plutonium was estimated by this method. This technique may be useful in future studies of the effect of chelating agents on plutonium in vitro.

BIOASSAY FOR PLUTONIUM

A review of current methods for bioassay of plutonium is given by Low-Beer[366]. Collection and initial handling of samples is included.

The history of the plutonium bioassay program at Los Alamos Scientific Laboratory from 1944 to 1972 was reviewed by Campbell[367] et al. Methods of urine sample collection, radiochemical separation, and counting are described.

A number of investigators have published methods for determination of plutonium in urine samples. In Japan, Hara[368] et al. used a method of ion exchange, and counted with a 2π gas flow counter. When plutonium is ingested or inhaled, fecal samples are collected for 3 to 5 days after intake. In France, Ventadour[369] et al. have a rapid method using a large surface area xenon-methane proportional counter having low background noise.

In India, Iyer and Kamath[370] have developed a rapid determination of uranium, thorium, plutonium, and americium sequentially in a single urine sample. The actinides are precipitated with $BiPO_4$ and the hydrolysis method used for separation.

In Russia, Mikhailova[371] et al. developed a method in which Pu(IV) as the cupferronate is separated from macro impurities by chloroform extraction from a nitric acid medium using zirconium carrier or no carrier. About 3 percent of the ^{239}Pu remains in the aqueous phase. After removal of the chloroform from the organic phase, the ^{239}Pu is precipitated with ammonia, using a lanthanum carrier and is counted after further processing. The plutonium can also be electro-precipitated. Golutvina[372] et al., in determining ^{239}Pu in the presence of enriched uranium, used a method based on boiling urine with nitric acid and hydrogen peroxide, concentration of isotopes by precipitation and subsequent extraction with precipitates of bismuth phosphate (^{239}Pu) and lantan fluoride (enriched uranium). The precipitates are mixed with a fluorescent compound and counted in a layer of hard scintillator. The efficacy of registration of alpha-particles comprises 90 to 95%.

In Germany, Schieferdecker[373] detected plutonium in urine and feces based on DEHPA extraction, and subsequent counting by alpha spectrometry. Activity amounts of 0.1 pCi can be determined.

In England, Popplewell and Stradling[374] developed a 2 to 3 hour determination for plutonium in urine. The procedure utilizes the fact that plutonium binds to transferrin in solutions of pH >6. This serum protein, M.W. 88,000, was filtered from aqueous solutions with appropriate membranes. Another rapid method was developed by Bates[375] et al. for determination of plutonium in urine within 4 hours of receipt, excluding counting time. The method consists of evaporating a 250-ml sample to dryness with nitric acid, baking at 550° C to remove organic matter, dissolution of the residue and removal of interfering condensed phosphates by boiling with a catalyst. The plutonium was isolated by an anion-exchange column and finally counting the plutonium on a tray in a low background counter.

In Italy, Camera[376] et al. described a method for the determination of alpha activity in urine by means of extraction, in a beaker, of radionuclides in a solution of triactylphosphine oxide (TOPO) on a base of Mitene 350/80 (polyethylene). Testa[377] et al. reported using wet mineralization, plutonium coprecipitation, a passage in an anion-exchange resin column, an electroplating procedure and alpha counting on an automatic solid state detector. They reported recovery of 78 ± 22%, with a sensitivity limit of 0.04 pCi/l or urine.

Campbell and McInroy[378] discussed the various aspects of handling human autopsy tissues and methods used for the determination of plutonium. It was pointed out that 5 mrem/y has been proposed as the maximum permissible level of plutonium for the general human population, which is equal to approximately three 1 μm particles of plutonium deposited per person per year and the analytical chemist is confronted with what appears to be an almost insurmountable task to detect this level of plutonium in man. The interlaboratory calibration of counting methods, using standards of ^{239}Pu, ^{238}Pu, and ^{236}Pu plated on stainless steel or samples of ashed beef bones, liver and lung spiked with ^{239}Pu, was discussed. Methods used for the chemical preparation of the ashed tissue samples for radiometric analysis were included.

Once analyzed the process of determining body content from urine data is just begun. Evaluation of excretion data by computer programs, models, and equations was reported by many investigators (Henle and Bramson[379], Heid[380,381] et al., Osanov[382,383] et al., Snyder[384], Nelson[385] and Beach[386]).

IN VIVO MEASUREMENT OF PLUTONIUM

The detection and measurement of plutonium in wounds was the subject of an article by Sharma and Somasundaram[387] in 1971. A thin NaI(Tl) scintillation detector was optimized for the detection of soft x-rays for use as a ^{239}Pu wound monitor. The resolution obtained for 17-keV x-rays was between 60 and 65%. Data on monitor sensitivity, background, minimum detectable ^{239}Pu activity and variation of the count rate in the plutonium channel with depth of plutonium source in a simulated human wound were presented.

Ohlenschlaeger and Fromhein[388] reported on the operational and technical features of a measuring system for the determination of radionuclides, particularly ^{239}Pu, in contaminated wounds. Special consideration was given to the integration of different detectors and devices into a mobile and portable measuring unit with central control. Experimental results gave proof of the system performance.

As in vivo determination of plutonium is only as good as the standardization of the systems used for its detection; a number of authors have reported on calibration of these systems.

An IAEA plutonium interlaboratory calibration experiment was described by Button[389] in 1973. Calibration factors for the in vivo measurement of ^{238}Pu and ^{239}Pu in human lungs as a function of body size were obtained using inhaled ^{51}Cr and ^{103}Pd particles as simulants. Optimum subject-detector geometry for the in vivo measurement of low-energy photon emitters in human lungs was determined. A mathematical model for americium and plutonium distribution in human lungs was developed. The performance of scintillation detectors inside a shielded whole-body counting room was evaluated.

Tomlinson[390] et al. reported in 1973 on chest wall tissue measurements for lung counting applications. Because the half-layer for 17-keV x-rays in tissue is only 6 to 7 mm, the effective thickness of tissue overlying the lungs was determined by ultrasonic measurements over the second, third and fourth rib.

In 1973, Swinth and Dean[391] discussed an intercalibration program consisting of tabulation of photon intensities for actinides, an intercomparison of counting systems using a standard consisting of either a source or a phantom, and counting of a subject or subjects who had been accidentally exposed to plutonium.

Tomlinson[392] et al. reviewed the basic, whole body counting program for the Mound Laboratory since December 1969. The phoswich detection system is described. Detection limits were given as a function of the subject's tissue thickness between the lungs and detectors. For a typical subject with an effective tissue thickness of 2.3 cm over the lungs, the system has a detection limit of 4 nCi.

Detection systems with various modifications of NaI(Tl) crystals and proportional counters have been reported by many investigators. The problems of detecting low-energy photons from plutonium deposited in the human body were discussed by these investigators and detectors modified to attempt to obtain direct plutonium measurement for assessing body burdens (Yaniv[393], Newton[394] et al., Ishihara[395] et al., Tomitani and Tanaka[396], Sharma[397] et al., Dolgirev[398] et al., Clemente[399], Boulay[400], Morsy[401], Loessner[402], and Newton[403] et al.).

Swinth[404] et al., Swinth and Ewins[405], Swinth[406], and Moldofsky[407] discuss in vivo plutonium measurements in tracheobronchial lymph nodes by means of intraesophogeal probes designed to minimize attenuation of the x-rays associated with plutonium.

MONITORING INSTRUMENTATION

As the primary hazard of plutonium to humans is inhalation of respirable aerosols and particulates, improvements and modifications in air filters and monitoring instrumentation have been continually under investigation.

Davis[408], Angel and Anderson[409], and Ettinger[410] et al. discussed the use of high-efficiency particulate air (HEPA) filters for plutonium particle and aerosol entrapment.

Plutonium monitoring instruments for laboratory use were reported by Binard[411], Hardy[412] et al., Nichols[413], Singh and Chugh[414], and Kikkawa[415] et al.

The instrument described by Nichols[413] was found to eliminate greater than 90% of the 2,250 dpm alpha natural background activity collected in order to detect 48 RCGS of soluble ^{239}Pu within 1-1/2 hours. The instrument utilized two ZnS scintillation detectors. It was believed that by using discriminators, lower concentrations of ^{239}Pu could be detected.

Singh and Chugh[414] reported using an alpha spectrometer technique using a silicon surface barrier detector. The system is capable of indicating alarm at a level of 8 MPC hours in the presence of natural activity.

Portable plutonium detectors for use in the field were discussed by Burton and Anastasi[416] who used a probe with a CaF(Eu) scintillator with a pulse height analytical capability to detect fissile material in the presence of background. The absolute sensitivity is $\sim$24 counts/min/$\mu g/m^2$ of ^{239}Pu on a uniformly contaminated surface with no overburden.

Cohen and Gundersen[417] discussed field experience with the mobile AEC measurement van.

The necessity of surveying large areas of land has required the development of instruments for aerial surveys. Stuart[418] reported on such a system, set to sense the 60-keV x-ray from ^{241}Am, a decay product of ^{241}Pu. The detector was mounted inside an Air Force helicopter and flown over known concentrations at the Nevada Test Site.

Boyns and Anderson[419] discussed an airborne spectrometer designed to search for ruptured plutonium capsules in event of an accident involving a space mission utilizing ^{238}Pu thermoelectric generators. A spectrometer capable of detecting ^{238}Pu in air below the maximum permissible concentration of 10^{-12} $\mu Ci/cm^3$ without interference from naturally occurring airborne alpha emitters was mounted in an Airborne Radiological Monitoring System (ARMS). Information was included on the aircraft positioning system, air sampling, and the procedures to be followed when actually using the spectromeeter.

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